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The Report of the Joint Task Force on Uranium Recycle Materials Processing

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THE REPORT OF THE
JOINT TASK FORCE ON
URANIUM RECYCLE MATERIALS PROCESSING

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Abstract

A Joint Task Force on Uranium Recycle Materials Processing was assembled by the Department of Energy (DOE) to study past and current practices relating to the processing of uranium recycle materials at DOE's Feed Materials Production Center (FMPC), Oak Ridge Y-12 Plant, and the DOE operations at the RMI Company. The DOE facilities providing the uranium recycle material and selected end users of the recycle material were reviewed in a cursory manner.

The Task Force determined that uranium recycle material produced by the DOE reprocessing sites will always contain trace levels of transuranics (e.g., plutonium and neptunium) and fission product (e.g., strontium and cesium) elements. However, the DOE processing sites such as the FMPC, Y-12 and RMI Company can safely handle and further process the recycle material if a clear understanding of the contaminant levels exists and available technology is utilized to assure environmental, safety, and health protection of both the plant worker and the general public. It was recognized that past practices regarding the processing of recycle materials could have been better (e.g., better understanding of contaminant levels in the feed material), however, from the data reviewed, the Task Force did not disclose any instance in which the environment, safety or health of plant workers or the public were jeopardized or compromised. It should be made clear that a lack of data hampered the Task Force throughout its efforts.

Irrespective of past practices, the Task Force judged that more attention should be given to the processing of uranium recycle material. The primary recommendation from this study is to develop formal specifications on maximum permissible levels of contaminants in feed materials. This work is already underway with an expected completion date of September 1985. Deficiencies in personnel/contamination control and environmental monitoring were confirmed by the Task Force; however, efforts were already underway to effect previously requested improvements. Additionally, recommendations were offered by the Task Force for a closer examination of selected recycle material workers at the Paducah and Portsmouth Gaseous Diffusion Plants due to unique processing operations at those DOE sites. This work is also underway.

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EXECUTIVE SUMMARY

Introduction

A DOE-wide Task Force was appointed on April 17, 1985, to examine the adequacy of past and current practices and procedures as they relate to the level of radioactive contaminants in uranium recycle materials handled at the Oak Ridge Y-12 Plant, the Feed Materials Production Center (FMPC) in Fernald, Ohio, and the RMI, Company in Ashtabula, Ohio. The source of the uranium recycle material to these plants is primarily from production reactor spent fuel discharges and subsequent reprocessing operations at the Savannah River Plant (SRP), Hanford, and the Idaho Chemical Processing Plant (ICPP). The reprocessing operation results in two primary streams. The more significant stream is the recovery of plutonium for the nuclear weapons program. The secondary stream is the remaining uranium stream which is slightly contaminated with trace quantities of transuranics (e.g., plutonium, neptunium) and fission products (e.g., technetium). It is this secondary stream that the Task Force studied and herein reports on.

The Task Force chose to fulfill its assignment by reviewing DOE uranium recycle operations from three different perspectives--the front end spent reactor fuel reprocessing sites (the SRP, Hanford, and the ICPP); the major intermediate recycle processing sites (the FMPC, RMI, and Y-12); and the end user or customer. Most of the Task Force effort was devoted to the intermediate processing sites. During the study, the Task Force effort was expanded to briefly address past operations that were conducted at the Paducah Feed Plant and the Portsmouth Oxide Conversion Facility since both sites processed some recycle materials until shutdown in 1977.

To assist the reader in understanding the breath of sites studied by the Task Force, a composite flow sheet showing the major flows of recycle material and the DOE sites involved in this study is presented in Figure 1.

MAJOR URANIUM RECYCLE FLOWS

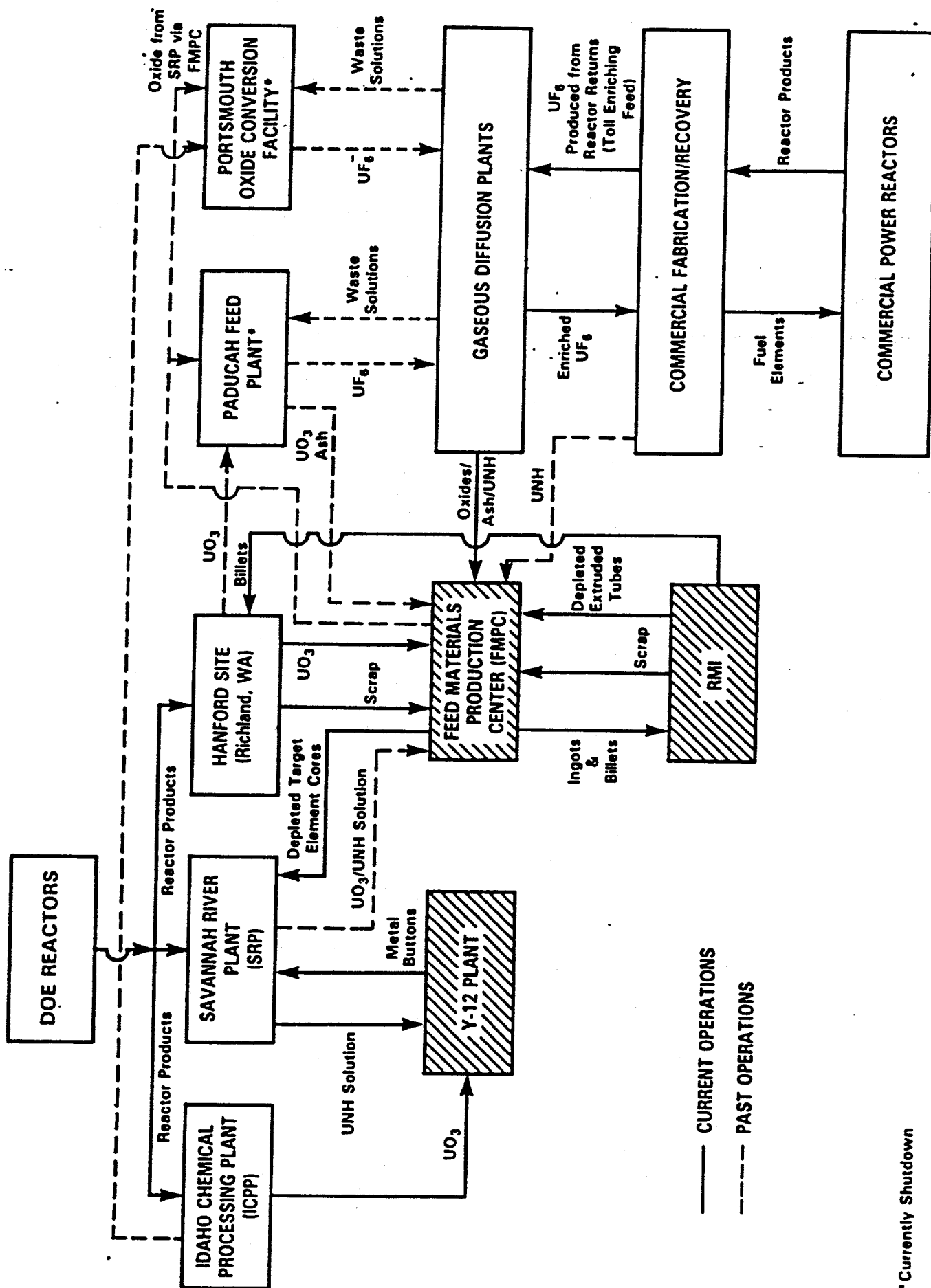


Figure 1
viii

Currently Shutdown

Findings

The Task Force found that the DOE spent fuel reprocessing sites have been safely recovering spent reactor fuel through proven chemical reprocessing schemes. However, all of the reprocessing schemes produce recycle material with trace quantities of transuranic and fission product elements. It is economically impracticable to produce recycle material from used nuclear fuel without some trace contaminants (transuranic and fission product elements) being present. It appears that the reprocessing sites are producing recycle material as free of contaminants as reasonably can be expected.

The recycle material that is produced by the reprocessing sites can be safely handled and further processed by the intermediate processing sites if a clear understanding of the contaminant levels exists and available technology (where needed) is utilized to assure environmental, safety and health protection of both the plant worker and the general public. Historically, the Task Force noted that a formal understanding of the contaminant levels was not always apparent as evidenced by the lack of formal specifications on maximum permissible contaminant levels between the reprocessing, intermediate, and customer sites. Such specifications are the primary recommendation from the Task Force. Although the intermediate processing sites may not have had a formal understanding of contaminant levels in recycle materials, sufficient information was available and/or developed to permit the further processing of recycle materials in a manner that did not jeopardize or compromise the environment, safety or health of the worker or the public as best can be judged by the Task Force. It should be made clear that only very limited information and data were available from historical records to: (1) characterize the level of contaminants received from the reprocessing sites; (2) examine the full extent of methods implemented to protect the worker's health and safety; (3) examine the flow of contaminants during processing operations; and (4) characterize the ultimate fate of the contaminants.

Irrespective of past practices, the Task Force judged that more attention regarding the processing of recycle materials is required to maintain better control of transuranic and fission product elements. It is essential that

personnel and contamination control programs fully recognize and reflect the levels of contaminants that may exist from time to time. As such, it may be sometimes necessary to implement a more rigid radiation protection program to assure adequate control over contaminants. The Task Force did not offer recommendations concerning personnel and contamination control and environmental monitoring since these were earlier addressed by special DOE reviews of the FMPC Health Physics and Environmental Programs. It should be understood that the degree of protection required is generally contingent upon the contaminant levels in the recycle material, and the environmental, safety and health protection practices must be established accordingly. Of interest, both the Health Physics and Environmental Monitoring Reviews documented many shortcomings, however, no evidence existed to indicate that worker health and safety were in jeopardy.

The Task Force expanded its efforts to cursorily review recycle material processing at the Paducah Feed Plant and the Portsmouth Oxide Conversion Facility. Both of these sites have received recycle materials from reprocessing sites and converted the recycle materials to uranium hexafluoride for feeding to the Gaseous Diffusion Plants. The Task Force judges that a closer examination of the past practices in the handling and processing of recycle material at both Paducah and Portsmouth is warranted, and recommendations in this regard are offered. In addition, the Task Force learned that quantities of recycle materials with undetermined levels of contaminants are being maintained by the gaseous diffusion plant complex. This dilemma is being further complicated by periodic receipts of uranium hexafluoride commercially produced from spent reactor fuel (primarily foreign) which contain trace levels of contaminants. Although feeding of this material to the gaseous diffusion plants has been done in the past, this is not a highly desirable operation due to the potential impacts on personnel safety, process productivity, and product purity of the gaseous diffusion plants. The Task Force offered a recommendation for the diffusion plant contractors to study this problem, outline available options, and provide a consensus recommendation to DOE.

Summary and Conclusions

Based on an analysis of the findings and observations of this report, the Task Force developed the following remarks.

Generic (Applies to All Sites)

- o A formal, technically sound, understood and accepted specification for maximum transuranic and fission product contaminants in uranium recycle material has probably never existed either within or between sites. Although most sites had their own "working" specification, there simply was no understanding and agreement on specifications for recycle material shipped to or from the DOE sites studied by the Task Force that had been agreed to, signed, and used for decision-making.
- o The Task Force found no reasons to propose a change or modification to the basic process flowsheets currently used at each of the plants reviewed. However, there does appear to be some instances where management attention is needed to improve environment, safety and health activities along with a stronger emphasis on "as low as reasonably achievable" (ALARA) concepts.

FMPC

- o The FMPC has not been required by DOE to maintain accountability records of transuranic and fission product elements in the quantities generally received by the FMPC. As such, the Task Force could not determine, with confidence, the quantity of contaminants that may have been received and processed at the FMPC. Only best estimates were available for the review.
- o Of all the plutonium estimated to have been received by the FMPC over the past 24 years (since plant startup), 50 percent of the plutonium was thought to have been contained in one shipment of Paducah Feed Plant ash in 1980. About 32 percent is believed to have been received from the Hanford site. The balance of the plutonium came from NFS-West Valley, the SRP, and other miscellaneous sources.

- o In June 1980, 22.5 metric tons of uranium ash from the Paducah Feed Plant was shipped to the FMPC for processing. This material was shipped to FMPC with DOE approval; however, it could not be established by the Task Force that DOE was formally aware of the plutonium levels in the ash. This recycle material, based on limited, non-homogeneous sampling, is estimated to have had plutonium levels ranging from 67 ppb to 7,757 ppb. In 1982, some of this material was processed and shipped to meet customer requirements. About 168 MTU of the remaining material (produced by diluting the original 22.5 MTU of Paducah Ash) currently remains at FMPC in the form of UO_3 with a maximum concentration of 43 ppb plutonium. Special precautions will need to be taken to process this material.
- o Overall, based on information made available, the Task Force found no evidence that DOE environmental, safety, and health guides had been exceeded nor was there any evidence that the health and safety of FMPC workers or the general public had been compromised due to operations involving recycle material. In addition, there was no evidence that the environment surrounding the FMPC had been adversely impacted from recycle operations. Nevertheless, deficiencies in the management control systems were noted which indicate a need for more environment, safety and health attention to uranium recycle processing operations.
- o The Task Force concluded that insufficient effort and attention was given to worker safety and radiation exposure control. For example, during routine operations the decision on whether an ingestion of radioactive material has taken or could take place rests with the worker. It did not appear to the Task Force that workers have had enough training and/or knowledge to intelligently make such decisions.
- o Routine processing of recycle materials containing less than 10 ppb plutonium can be accomplished with existing administrative and radiation protection practices. This is true since uranium is the dominant radionuclide for health protection purposes at a plutonium concentration less than 10 ppb. Most of the radiation protection practices are those that should be

implemented to support uranium operations. The requirement should be to assure that uranium is always the controlling nuclide for any processing operation regardless of the contaminants that may be in the feed material.

- o Deficiencies in the FMPC Health Physics and Environmental Programs were noted by the Task Force. However, recent special reviews have pointed out these deficiencies, and as such, the Task Force chose not to repeat those same findings and recommendations previously observed.

RMI

- o Depleted and slightly enriched uranium ingots from FMPC are extruded into tubes at RMI and then shipped to Hanford or back to the FMPC for final work. No contaminants are added to or removed from the uranium being worked at RMI. RMI does, however, convert uranium turnings to an oxide form prior to being returned to FMPC.
- o RMI is not equipped to perform sampling analysis for transuranic or fission products. RMI analyses are generally accomplished by the FMPC. In addition, outside laboratories and expertise are available to RMI.
- o A recent risk and vulnerability study of RMI outlined several improvements that would benefit recycle material processing operations at RMI.

Y-12

- o Limited data exists at Y-12 on the transuranic and fission product content of recycle material receipts, processing streams, and product streams. As in the case of the FMPC, Y-12 has not been required to maintain accountability data on plutonium, other transuranic elements, or fission products.
- o An exposure assessment of recycle workers indicate these workers have an external exposure 1.2-1.6 times the exposure of Y-12 workers handling unirradiated (virgin) uranium materials. The internal dose to the recycle

workers were calculated by Y-12 to be 0.019 rem per year (committed dose to the bone) per employee. The Task Force agreed with Y-12 that these exposures do not represent a significant health or safety risk to the recycle workers. These calculations were based on a review of employee exposure records.

- o Based on limited sampling since 1977, the Y-12 staff has noted a buildup of fission products in both the liquid and solid waste streams as a result of processing recycle material. It appears that this waste stream buildup has taken place as the fission product concentration in Y-12 product has decreased. The waste streams were previously routed to the S-3 Ponds; however, the S-3 Ponds have been closed, and a closure plan has been developed by the Y-12 Plant in conjunction with local, state and federal agencies.

Paducah Feed Plant

- o The Paducah Feed Plant (shutdown in 1977) maintains an inventory of about 0.8 MTU as UO_3 and 1.2 MTU as miscellaneous solids processed from recycle material. In addition, about 1.3 MTU as ash remains from the cleanout of the Paducah Feed Plant. Plans for disposition of this recycle material have not been formulated. Such a plan is a recommendation of the Task Force.

Portsmouth Oxide Conversion Facility

- o This facility (shutdown since about 1977) was used to convert high assay uranium recycle material to UF_6 for feed into the gaseous diffusion plant. An estimated several thousand 5-inch diameter cans possibly containing some level of transuranic and fission product contaminants are in storage at the site. Non- UF_6 material is being shipped to Y-12 (high assay) or the FMPC (low assay) on an as-needed basis.

Gaseous Diffusion Plants (GDP)

- o It is estimated that the three GDPs currently have about 613 MTU as UF₆ that was commercially processed from recycle material. The assay of this material generally is about 0.8 percent U-235, but some material goes as high as 86 percent U-235. In recent years, the problems associated with feeding uranium recycle material to the GDPs have been studied. The technology for removing transuranics (primarily Np-237) and fission products (primarily Tc-99) from incoming feed is judged to be available; however, this technology has not been demonstrated on a production level. Action on this technology is currently receiving low priority attention by DOE due to economic considerations.

Recommendations

The Uranium Recycle Task Force offers the following recommendations for DOE's consideration:

Generic (Applies to All Sites)

Mutually agreeable and technically sound transuranic and fission product element specifications should be established between shipper and receiver for all recycle material shipped to and from all DOE sites handling recycle material. The specifications should be agreed upon and signed by all involved contractors, and approved by the appropriate DOE field offices. These specifications should be provided to all affected plants.

To support the recycle material specifications noted above, a documented technical basis should be prepared for those radioactive contaminants (transuranics and fission products) covered by the specification. A justification for each limit should be given. To implement this recommendation, DOE established a multi-contractor Specifications Task Group to develop these specifications under the auspices of the Uranium Recycle Task Force.

FMPC

FMPC management should:

1. Continue to recognize the 10 ppb Pu (on a uranium basis) specification (upper limit) on recycle materials until the results of the Specifications Task Group noted above completes its work. The 10 ppb Pu guide is based on the rationale given on pages 16-19 of this report.
2. Regarding any remaining (especially the 168 MTU from the Paducah Ash) or future receipts of recycle material at FMPC, formalize the operating and environment, safety and health procedures; handling methods; and analyses to be utilized in future processing operations which will clearly demonstrate that no adverse environmental, safety or health effects will occur from recycle material processing. To the extent possible, engineering controls are preferred over administrative controls. The processing of recycle material in concentrations greater than 10 ppb plutonium will require special DOE-ORO approval if it is necessary to process recycle material prior to completion of the Specification Task Group work.
3. Comprehensively review the analytical and radiochemical procedures and control programs for determining transuranic and fission product elements in the incoming, in-process, and product streams as well as in environmental samples. Procedures and equipment should be upgraded to state-of-the-art technology and be capable of identifying individual radionuclides, e.g., Pu-238 and Pu-239 to the extent possible:
4. Obtain expert advice and guidance in establishing a bioassay program including in-vivo (whole body) counting for FMPC workers. To the extent feasible, the program should include the measurement of transuranic and fission product burdens. Until such time as an onsite capability can be established, FMPC should obtain bioassay data (if this has not already

been accomplished) on those workers involved in recycle material operations. In addition, those recycle workers with the highest potential of lung internal deposits of plutonium should be sent to facilities with in vivo counters for measurement.

5. Review the FMPC training program for operators, supervisors and maintenance personnel to assure compliance with DOE Order 5480.1A, Chapter V, paragraph 8. In addition, FMPC should review operating and environment, safety and health (ES&H) procedures to assure these procedures are current, readily available to and used by personnel, periodically reviewed/updated as necessary, and have been properly reviewed by the ES&H staff.

RMI

RMI management should:

1. Recognize and support the FMPC 10 ppb Pu (on a uranium basis) specification on DOE recycle materials until the results of the Specifications Task Group noted above completes its work. The 10 ppb Pu guide is based on the rationale presented on pages 16-19 of this report.
2. Implement a reasonable program of sampling and analyses on incoming, in-process and outgoing batches of recycle material. RMI efforts should include process residues and waste streams, as appropriate.
3. Include all uranium handling workers in the RMI bioassay program if this has not already been accomplished. To the extent feasible, the bioassay program should include transuranic and fission product measurements.

Y-12

Y-12 management should:

Review radiochemical procedures and control programs associated with the analysis of uranium recycle processing operations.

Paducah Feed Plant

Paducah management, in conjunction with DOE, should:

Conduct an exposure assessment (to transuranics and fission products) for those workers involved in the processing of recycle materials at the Paducah Feed Plant.

Portsmouth Oxide Conversion Facility

Portsmouth management, in conjunction with DOE, should:

Conduct an exposure assessment (to transuranics and fission products) for those oxide conversion facility workers involved in the processing of recycled materials.

DOE Gaseous Diffusion Plants (Oak Ridge, Paducah and Portsmouth)

GDP contractor management should:

Provide DOE with a report on the options available and recommendations for a safe, technically sound, and feasible method for disposition of current inventories and future GDP receipts of uranium recycle material (UF_6 and non- UF_6). The report should include, as a minimum, (a) an assessment of the problems presented by recycle materials on the GDP complex, (b) available disposition options, and (c) a recommended course of action to DOE. DOE will be responsible for a final decision in this matter.

I. INTRODUCTION

A DOE Task Force was appointed by the Manager, DOE Oak Ridge Operations Office and Don Ofte, Principal Deputy Assistant Secretary for Defense Programs, on April 17, 1985, to undertake an evaluation of operations involving radioactive contaminants in uranium recycle materials processed at the Oak Ridge Y-12 Plant, the Feed Materials Production Center (FMPC) near Fernald, Ohio, and the RMI, Company in Ashtabula, Ohio. (See Figure 1). The major driving force that resulted in the establishment of this Task Force was a recent inquiry regarding a FMPC processing campaign in 1982 of recycle material containing unusually high levels of plutonium (estimated maximum plutonium-239 concentration of 7,757 parts per billion).

For purposes of this report recycle material is defined to be material that has been recovered by chemical processing from reactor fuel which was previously irradiated in a reactor. The source of the recycle material is the DOE production, naval, and research and test reactor spent fuel discharges and the uranium recovered in subsequent reprocessing operations at the Savannah River, Hanford, and Idaho Chemical Processing Plants. In addition, some recycle material has been received from commercial sources. The role of Y-12, FMPC and RMI is to assist in the processing of uranium material into finished uranium metal of the proper enrichment and shape for manufacture into fuel and target elements for the DOE production reactors.

The Task Force Charter centered around the following four assignments:

- (1) Review past and present practices, procedures, and policies in the handling and processing of recycle materials at FMPC;
- (2) Assess the adequacy of these practices, procedures, and policies as they relate to radioactive contaminants in uranium recycle materials processed at DOE facilities;

(3) Consider current levels of radioactive contaminants from a standpoint of:

(a) technically achievable levels, and

(b) potential health and environmental impacts;

(4) Consider each site's

(a) analytical capabilities to determine levels of radioactive contaminants, including mandatory limits (if established); and

(b) upgradings which might or should be required for future operations.

The Task Force performed its mission by conducting onsite reviews of the Y-12 and the FMPC sites; gathering and interpreting information from other DOE sites handling recycle material; and performing independent review and analysis of pertinent material.

II. FINDINGS AND TASK FORCE OBSERVATIONS

A. The Feed Materials Production Center (FMPC) and RMI Company

1. Description of Physical Plants

a. General Overview - FMPC

The FMPC is located near Fernald, Ohio. It is a large scale integrated facility for the processing of various uranium compounds to produce uranium metal for use as nuclear weapons parts and in the fabrication of fuel core and target elements for production reactors. NLO, Inc. operates the FMPC under prime contract with the DOE (see Figure 2).

A wide variety of chemical and metallurgical process steps are utilized at the FMPC for the conversion of uranium materials to machined uranium ingots and billets for extrusion into tubular form for fabricating fuel cores and target fuel elements. These FMPC products are used in defense programs of the Department of Energy (DOE).

The principal product from FMPC operations is uranium metal in various physical forms having several standard isotopic assays. Most of the FMPC production stream metal is cast into ingots, center-drilled, and surface-machined for extrusion into tubes on the DOE extrusion press facilities located at the RMI Company in Ashtabula, Ohio. Some extrusions are returned to the FMPC where tube blanks undergo heat treating and fabrication into target elements for DOE reactors at the Savannah River Plant (SRP), in Aiken, South Carolina. Other extruded material is further processed into fuel billets, via an upset forge operation

FEED MATERIALS PRODUCTION CENTER

PROCESS FLOW

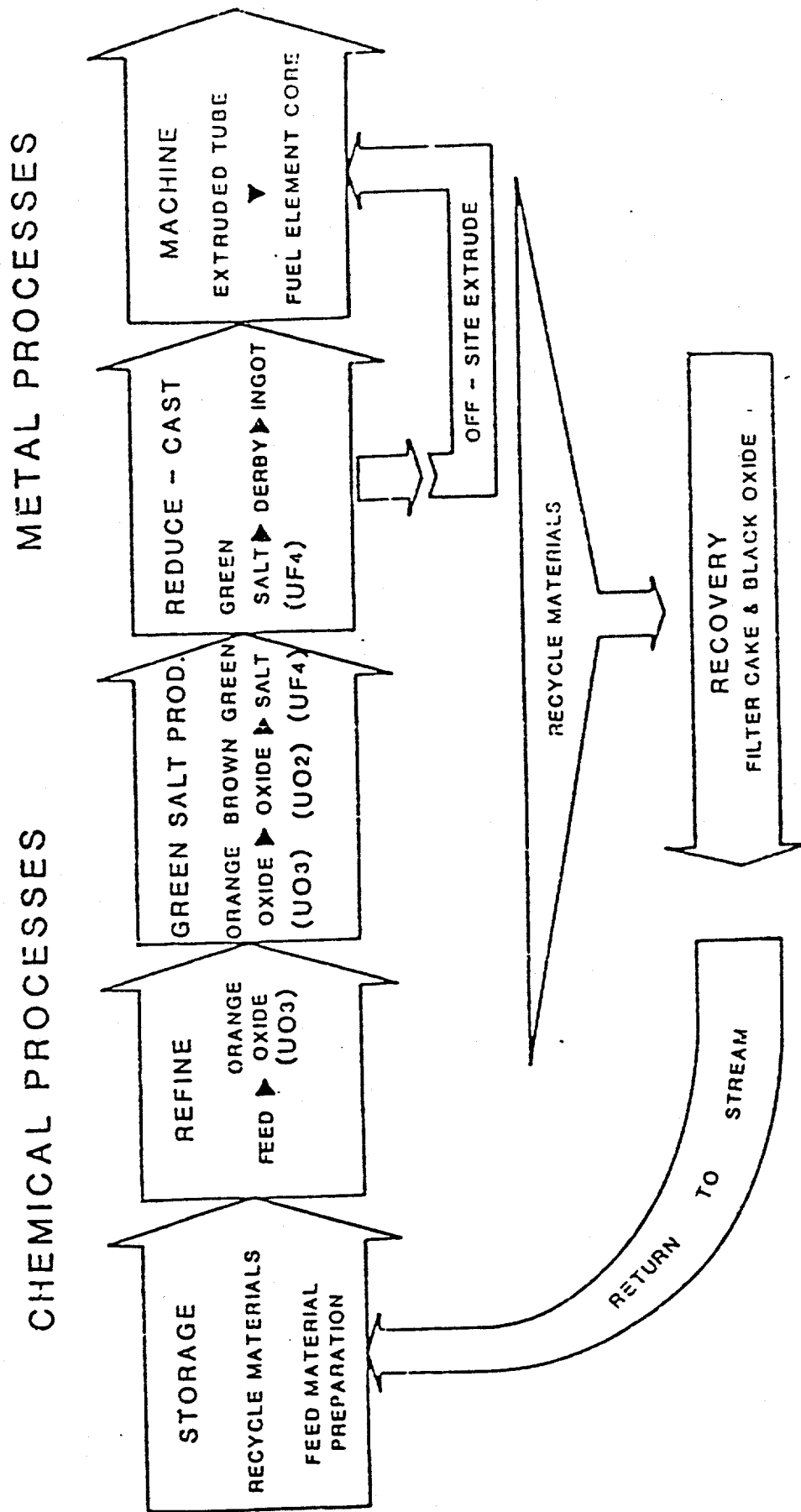


Figure 2
-4-

at RMI, for shipment to Hanford. Both fuel cores and target elements are used in DOE reactors for the generation of byproduct electricity (only at Hanford) and the production of plutonium.

The FMPC plays an important role in Defense Programs. FMPC products contribute to the DOE production reactors at the Hanford site near Richland, Washington, and Savannah River, South Carolina. In addition, some FMPC product is used in other defense production activities such as in the nuclear weapons program.

In the past, uranium oxides (UO_3 and U_3O_8) have been produced in the FMPC chemical operations to provide a feed material for DOE gaseous diffusion plants at Paducah, Kentucky, Oak Ridge, Tennessee, and Piketon, Ohio. This work has not been conducted in many years and is not likely to occur again in the foreseeable future.

RMI Company

The RMI Company Extrusion Plant is a privately owned facility utilizing a DOE-owned 3,850-ton Loewy extrusion press and support equipment for the purpose of converting DOE uranium metal from one shape to another via hot extrusion. Depleted and slightly enriched uranium ingots from the FMPC are extruded into tubes at RMI. The depleted uranium tubes are returned to the FMPC for final machining. (At the FMPC, depleted uranium tubes are cut into pieces and machined to form hollow cores, which are then shipped to the SRP for use in the production reactors.) Slightly enriched uranium ingots are fabricated into billets at RMI. These billets are shipped to Hanford where they are co-extruded with zirconium to form fuel elements for the N-Reactor located at the Hanford site.

The RMI Extrusion Plant (with the permission of DOE) performs extrusions on the DOE press for the private sector, including contractors to the Department of Defense. Depleted uranium is extruded for the private sector under the authority of a Nuclear Regulatory Commission (NRC) license. Copper, zirconium and other metals have also been extruded.

b. Uranium Recovery and Processing - FMPC

Large-scale chemical recovery operations at the FMPC consist of dissolving virgin and recycle uranium materials in nitric acid to produce a uranyl nitrate hexahydrate (UNH) feed solution for solvent extraction purification. Purified UNH solution is concentrated by evaporation and then thermally denitrated to uranium trioxide (UO_3), commonly called orange oxide. Orange oxide is converted to uranium tetrafluoride (UF_4), commonly called green salt, for reduction to metal. Scrap uranium materials generated in FMPC operations and those received from offsite are refined through chemical processing. Small-scale facilities exist for performing similar chemical process operations for enriched uranium assaying up to 20 percent U-235. In 1984, facilities were placed in operation for converting UF_6 to UF_4 green salt for subsequent reduction to derby metal.

Metal processing steps begin with the conversion of green salt to elemental uranium metal (derbies) by reducing UF_4 with magnesium metal. Metallic uranium scrap, recycled from subsequent fabrication operations, are combined with derby metal and melted in a graphite crucible inside an evacuated, induction heated furnace. The melt is bottom-poured to a preheated graphite mold to form ingots, varying in weight, size and shape according to their ultimate use. Cast ingots are then machined for extrusion into tubes at RMI. Most extruded tubes are returned to the FMPC for heat treating and final machining operations to produce target element cores for SRP. Some extrusions are processed at RMI to produce billets for co-extrusion at Hanford and other appropriate sites. Figure 3 provides a schematic diagram of the FMPC process.

SCHEMATIC DIAGRAM OF THE FMPC PROCESS

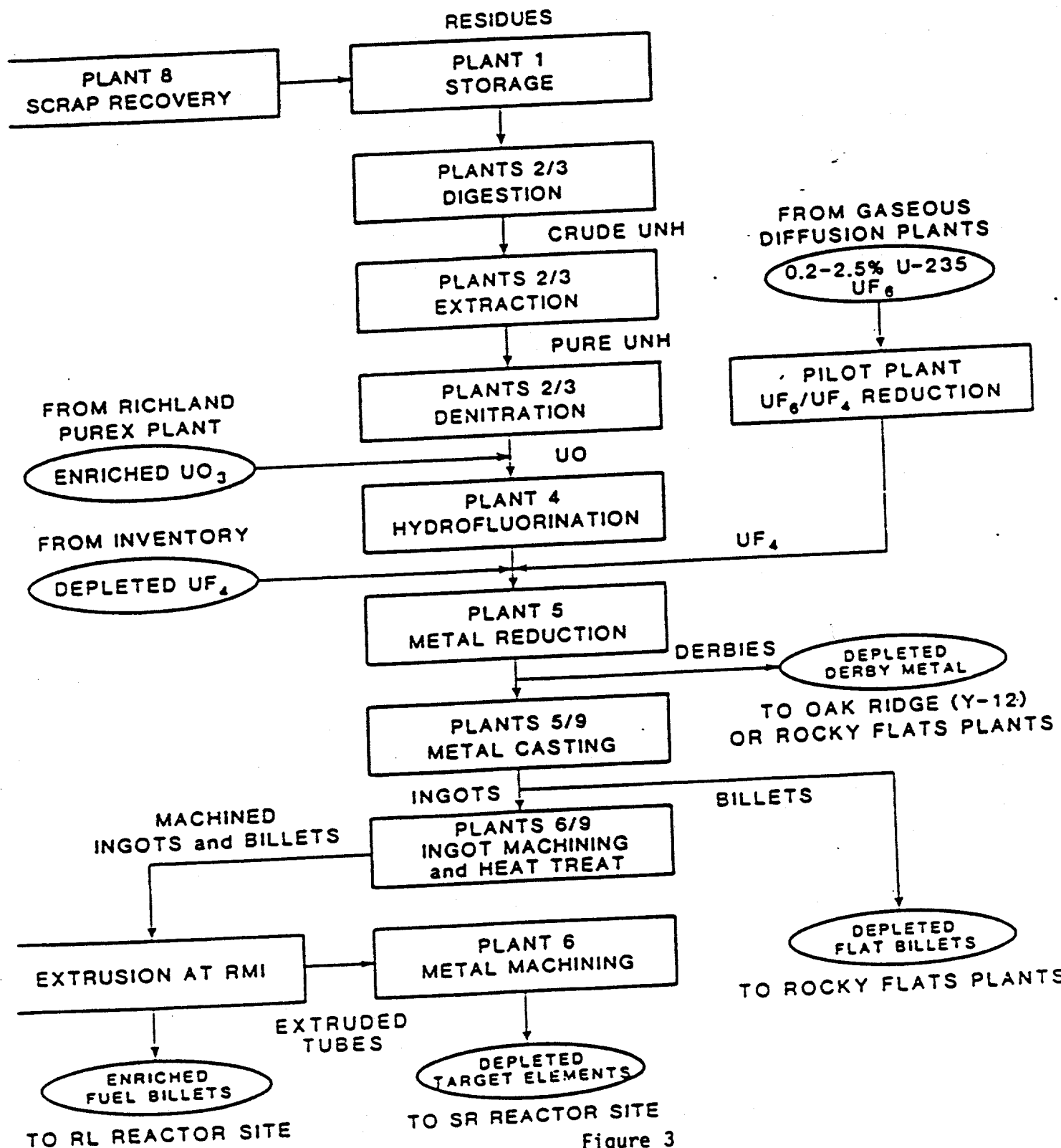


Figure 3

RMI

The RMI facility performs extrusions for DOE. Uranium bearing wastes, residues and reject materials generated by RMI in this effort are packaged for shipment to the FMPC for recovery/disposal. RMI does not have facilities for the recovery or processing of waste or side streams. In addition, the RMI facility is not equipped to perform radioisotope sampling analysis for materials processed at the site. However, samples are routinely taken of the process residues and sent to FMPC for analysis for nuclear materials accountability purposes.

Task Force Observation

As a result of the role played by RMI in recycle materials processing, little information was available to analyze. The Task Force was made aware of a recently released study conducted by DOE (RMI Extrusion Plant Vulnerabilities and Risk Assessment Concentrating on Environmental, Safety, Health and Quality Assurance Program Aspects) which pointed out deficiencies in some of RMI's operations. Based on the information available to the Task Force, the suggested improvements to the RMI operations would improve processing operations involving recycle materials. Of interest, the DOE assessment team found no serious deficiencies that would require immediate shutdown of any RMI operation. Also, the team identified no serious non-compliance situations with respect to DOE Orders or Federal or State law. However, the team noted that the lack of data limited their ability to definitely state that there were no violations of environmental or worker health protection criteria. Although 26 recommendations were offered in the report, those that were requested to receive the greatest and most immediate attention were as follows:

1. *Completion of planned workplace sampling for smoke and fume concentrations.*

2. *Implementation of the current hazardous waste storage/disposal policy.*
3. *Assessment of compliance status with respect to the National Emission Standard for Hazardous Air Pollutants.*
4. *Upgrading of the process wastewater treatment system.*
5. *Additional data collection of the status of onsite and offsite uranium contamination of soils.*
6. *Establishment of a full-time quality assurance position.*

2. Sources of Uranium - FMPC

Over the past 24 years, the FMPC has received about 504 million kilograms uranium (kg U) as feed material in all material forms (UO_3 , UNH solution, UO_2 , U_3O_8 , ash, etc.). Of this total, it is estimated that 1.5 percent (or 7.5 million kg U) consisted of recycled material. A summary of these recycled materials and the estimated plutonium-239 content are given in Table 1. Table 2 provides an insight to the estimated quantity of plutonium contained in the total of all recycled materials. The table shows that almost 56 percent of the plutonium received by the FMPC came from the Paducah Feed Plant (which originally received it from Hanford). Looking at this quantity differently, 56 percent of the plutonium was contained in 4 percent of the recycle materials.

Task Force Observation

The FMPC has not been required to maintain an accounting of the quantities of plutonium received at the FMPC. The plutonium data presented in Tables 1 and 2 are best estimates of what might have been received. These data are based on very limited shipper/receiver information which were reconstructed from historical shipping documents.

Table 1
Estimated Recycled Materials Received by the FMPC

(1961 - 1985)

<u>Material Type</u>	<u>Kg U</u>	<u>Pu-239 Grams</u>	<u>Average Pu-239 ppb (U Basis)</u>
Orange Oxide (UO ₃)	6,111,975	17.407	2.848
Uranyl Nitrate Hexahydrate (UNH)	699,093	4.706	6.732
Incinerator Ash, Tower Ash, and Black Oxide (U ₃ O ₈)	310,819	0.228	0.734
Brown Oxide (UO ₂)	39,177	0.010	0.255
Feed Plant Ash	22,529	25.290	1122.553
Miscellaneous Scrap from Paducah (1975-1976)	<u>290,742</u>	<u>2.610</u>	8.977
Total Recycled Materials	<u>7,474,335</u>	<u>50.251</u>	<u>6.723</u>

NOTE:

Although the data shown in this table are given to several significant figures, these data are only "best" estimates of what might have been received. These data are based on reconstructed, historical information. (See Task Force observation on the previous page.)

Table 2
Sources of Plutonium Received by FMPC

(1961 - 1985)

<u>Source</u>	<u>Kg. Uranium</u>	<u>Gm. Plutonium</u>	<u>Percent of Total Pu</u>
Paducah Feed Plant*	313,271	27.9	55.5
Hanford Recycle**	5,589,591	15.968	31.8
NFS, West Valley	617,877	4.160	8.3
Savannah River Plant	669,026	1.996	4.0
All Other Sources	<u>284,570</u>	<u>0.227</u>	<u>0.4</u>
Totals	7,474,335	50.251	100

*Based on FMPC data.

**A part of this material was UO₃ received from Hanford after intermediate storage at Paducah.

NOTE:

See the note in Table 1 regarding the accuracy of this data.

RMI

As stated earlier, all DOE uranium metal received by RMI is shipped from the FMPC as ingots or billets for extrusion. The extruded products go to Hanford or FMPC. No radioactive contaminants are added to or removed from the uranium being worked at RMI. To avoid potential pyrophoric problems, the uranium metal scrap is converted to an oxide prior to being returned to FMPC. Bulk scrap is returned as is.

3. Standards/Specifications

There are inconsistencies regarding the maximum allowable transuranic and fission product content in recycle material processed by the FMPC. The following illustrate this point:

- o FMPC advised that "Internal NLO [the operating contractor of the FMPC] memos established 3,000 disintegrations per minute (dpm) plutonium and neptunium per gram of uranium (roughly 20 parts per billion [ppb]) as a target for maximum TRU [transuranic] content in materials handled at the FMPC. This has not been a standard or acceptance criterion. On a stream-average basis, only the Paducah Feed Plant Ash received in 1980 exceeded this level. Two samples from Paducah Scrap received in 1976 exceeded this level, although the average of all samples was well below the target TRU level." FMPC does not have fission product specifications for incoming recycle materials.

Task Force Observation

The FMPC advised the Task Force that the internal NLO memos quoted above were based on a DOE directive contained in a letter to the FMPC dated August 16, 1976 (discussed below).

- o During 1971, the Atomic Energy Commission (now DOE) published specifications for the commercial delivery of uranium oxides, UNH crystals and UNH solutions to AEC facilities for conversion to uranium hexafluoride (UF₆). An applicable page from these specifications is shown in Table 3. A limit of 15,000 dpm/g U alpha activity was established.
- o By memorandum dated November 21, 1975, W. D. Sandberg (SRO) to H. D. Fletcher (ORO), SRO indicated that the Savannah River Plant had been operating on a maximum alpha activity specification from all transuranic elements of 1,500 dpm per gram of total uranium, not the 15,000 dpm/g total uranium specified by the AEC. This specification applied to the low enriched UNH solution delivered to the FMPC. This specification change had been transmitted verbally by an employee of the Portsmouth Gaseous Diffusion Plant to DuPont at the SRP. In this same memorandum, SRO explained the difficulty being experienced at the SRP in attaining the 1,500 dpm/g U specification, and a reevaluation of the 1,500 dpm/g U specification was requested. After review, Fletcher responded to Sandberg on August 16, 1976 stating that a specification of 3,000 dpm/g total uranium should be applied to the UNH solution supplied by the SRP to the FMPC. It was further stated that alpha contamination, and therefore transuranics, lower than 3,000 dpm/g uranium should be supplied on a "best effort" basis. No reference was made to fission product specifications.
- o Applicable portions of the current specifications for the Hanford recycle uranium as a UO₃ product are as follows:

Actinides

- | | |
|----------------|---------------------|
| a) Uranium-235 | <1.0 wt% in uranium |
| b) Plutonium | <10 ppbp uranium |
| c) Thorium | <750 ppmp uranium* |

*Values are based on a lot composite sample of 10 containers.

Table 3

AEC Specifications

Item	Numerical Value for Each Chemical Form		
	Oxides* (Either UO_2 , UO_3 or U_3O_8)	Uranyl Nitrate Crystals* ($\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$)	Uranyl Nitrate Solution ($\text{UO}_2(\text{NO}_3)_2 + \text{H}_2\text{O}$)
Maximum total gamma activity from fission products and uranium-237 as percent of gamma activity of aged natural uranium and as measured in a high-pressure ionization chamber (Drawing D-AWM-8796 of Nuclear Division, Union Carbide Corp.):			
(a) provided that no less than 75% of the total fission product gamma activity is due to ruthenium isotopes	100	200	200
(b) provided that less than 75% of total fission product gamma activity is due to ruthenium isotopes	100	100	100
Maximum total beta activity from fission products as percent of the beta activity of aged natural uranium:			
(a) provided that no less than 75% of the total fission product beta activity is due to ruthenium isotopes	100	200	200
(b) provided that less than 75% of the total fission product beta activity is due to ruthenium isotopes	100	100	100
Maximum alpha activity from all transuranic elements in disintegrations per minute per gram of total uranium	15,000	15,000	15,000

*Uranium oxides and uranyl nitrate crystals shall be free-flowing powders.

Fission Products

- | | |
|----------------------------------------------|-----------------------------------------------------------------------|
| a) $^{95}\text{ZrNb}$ | $<15 \mu\text{Ci/lb uranium}^*$
$<10 \mu\text{Ci/lb uranium}^{**}$ |
| b) ^{103}Ru and $^{106}\text{RuRh}$ | $<50 \mu\text{Ci/lb uranium}^*$
$<25 \mu\text{Ci/lb uranium}^{**}$ |
| c) All others excluding ^{99}Tc | $<2 \mu\text{Ci/lb uranium}^*$
$<0.5 \mu\text{Ci/lb uranium}^{**}$ |

*Values are based on a lot composite sample of 10 containers.

**Values apply to a 10-lot average.

Note: ppbp is defined as parts plutonium per billion parts uranium.
ppmp is defined as parts thorium per million parts uranium.

While the Hanford UO_3 product specifications are used as a working document by both the FMPC and Hanford, no written agreement exists between the two sites that formalize these specifications.

- o In a March 26, 1985, internal FMPC memorandum, it is stated that a plutonium-239 concentration of 10 ppb or less (uranium basis) was such that plutonium and other transuranic elements in recycle uranium would not require special control efforts at the FMPC. In the memorandum FMPC established this 10 ppb limit to be equivalent to the 1,500 alpha dpm/g U. The memorandum stated that additional health protection controls would be required during processing recycled uranium containing plutonium concentrations between 10-50 ppb, and special training and handling procedures would be prudent during processing of concentrations above 50 ppb. The memorandum went on to state that at a maximum plutonium concentration of 10 ppb, controls for exposure to airborne material can be based on the uranium activity. In an April 4, 1985 letter to FMPC plant management, DOE imposed a 10 ppb plutonium upper limit unless otherwise specifically approved.

Task Force Observation

The foregoing illustrates that a generally accepted specification for the maximum level of transuranic and fission product elements in recycle material has not existed. Table 4 summarizes the various specifications that were (might have been) used by the FMPC.

Except for very few data related to effluent monitoring (see Table 7) and sediment concentrations (Tables 9 and 10), very little information is available on Tc-99. In fact, data on fission product elements have not been generated to any significant degree. The lack of data and information on fission products could not be explained by the Task Force except perhaps the FMPC did not recognize a need for such data during this time period.

The Task Force judged that it would be prudent to determine the basis for using a specification of 10 ppb plutonium on a uranium basis for transuranic radionuclides in recycle materials. The calculations shown below support the possibility of having developed this specification. These calculations were performed by the Task Force and should be reviewed in the context of a "first cut-general analysis" with due regard to the assumptions (or lack thereof) used by the Task Force.

Specific Activities:

Natural Uranium = 1.5×10^6 dpm/g Unat

Plutonium-239 at a mass concentration of 10 ppb (U basis) would be 10 nanograms Pu-239 per g U which is equal to 135×10^3 dpm/g U

At this concentration (10 ppb Pu-239) in natural uranium, the alpha activity ratio is

Table 4

Plutonium Specifications and Source - Summary

<u>Specification</u>	<u>Source</u>
Maximum Alpha Activity from All Transuranic Elements: 15,000 dpm	AEC
Hanford UO ₃ Product: Pu <10 ppb	Hanford
Maximum Alpha Activity from All Transuranic Elements: 1,500 dpm	DOE - Savannah River
Maximum Alpha Activity from All Transuranic Elements: 3,000 dpm	DOE - Oak Ridge
Pu-239 Less Than 10 ppb	DOE - Oak Ridge and FMPC

$$\frac{10 \text{ Ng Pu-239}}{1 \text{ g Unat}} = \frac{1.35 \times 10^3 \text{ dpm}}{1.5 \times 10^6 \text{ dpm}} = 0.9 \times 10^{-3} = 0.00090 \text{ or } 0.9/1000,$$

and 0.9/1000 is approximately equal to the alpha activity ratio of 1/1000

DOE Concentration Guides

DOE Order 5480.1A, Chapter XI - Requirements for Radiation Protection provides DOE concentration guides for radionuclides in air and water above natural background. Attachment XI-1, pages 8 and 12 shows the following data by air:

Pu-239 (soluble) = $2 \times 10^{-12} \mu\text{Ci/ml}$ which equates to about 4.4 dpm/m³ air (alpha) for controlled areas. Same value for Pu-238 (soluble).

Pu-239 (insoluble) = $4 \times 10^{-11} \mu\text{Ci/ml}$ which equates to about 88 dpm/m³ (alpha) for controlled areas. Value for Pu-238 (insoluble) is 66 dpm/m³.

Natural Uranium (insoluble) = $6 \times 10^{-11} \mu\text{Ci/ml}$ which equates to about 133 dpm/m³ (alpha) for controlled areas.

Natural Uranium (soluble) = $7 \times 10^{-11} \mu\text{Ci/ml}$ which equates to about 155 dpm/m³ (alpha) for controlled areas.

U-235 (insoluble) = $1 \times 10^{-10} \mu\text{Ci/ml}$ which equates to 222 dpm/m³.

Now, if the Pu-239 (soluble) activity is maintained at the alpha activity ratio relative to uranium as calculated above (1/1000), the contribution to the total alpha activity from the Pu-239 (soluble) can be calculated to be 3.5 percent of the DOE concentration guide for Pu-239 as shown below.

$$\frac{154 \text{ dpm U}}{\text{m}^3} \left| \frac{1}{1000} \right| \frac{\text{m}^3}{4.4 \text{ dpm Pu-239}} = 0.035 = 3.5\%$$

Thus, keeping the Pu-239 activity in the air to 1/1000 of the concentration guide for uranium would control the Pu-239 air concentration to 3.5% of the DOE concentration guide for soluble Pu-239.

Similar calculations at various concentrations of Pu-239 yields comparative percentages, as shown below.

<u>Pu-239 (soluble) Concentration</u>		<u>% Pu-239 of Concentration Guide</u>
<u>dpm (U basis)</u>	<u>ppb (U basis)</u>	
1,350	10	3.5
2,700	20	7.0
4,050	30	10.5
5,400	40	14.0
6,750	50	17.5
13,500	100	35.0

The Task Force judges the above calculations to be conservative in that:

1. The Pu-239 is considered to be soluble (in man, not chemical form).
2. All the Pu-239 is assumed to be optimum in particle size for inhalation and retention in the lungs.

However, the calculations use Pu-239 which is non-conservative because its specific activity is about 1/270 that for Pu-238. It is important to know both the amounts and kinds of transuranic radionuclides in the uranium.

RMI

Specifications for transuranic and fission product elements in incoming material (ingots and billets) and outgoing extrusions do not exist.

4. Processing of Recycle Material - FMPC

There is evidence that the FMPC has been aware of the need to consider and evaluate the environmental, safety and health impacts from processing recycle materials. The earliest documentation provided to the Task Force was a March 25, 1965 study that was based primarily on data provided by the Hanford site and personal discussions between Hanford, SRP, and the FMPC. Although the conclusion reached by the FMPC staff in this document is that "...we do not have any new problems.", there were several uncertainties surrounding the characterization of recycle materials from the generating sites.

A followup study performed by the FMPC and reported on September 16, 1965 was done to determine if new health or safety considerations would be necessary for the processing of recycle material. The overall conclusion from the followup study indicated that "...the processing of current recycle material does not require any health or safety considerations other than those made for normal [virgin] uranium."

Another report regarding recycle material processing made available to the Task Force was an environmental assessment issued by the FMPC on November 11, 1975 (NLCO-1130). The assessment indicated that "Most of the trans-uranics in liquid wastes will be precipitated with a great mass of residue and be discarded in onsite storage pits along with residues that are transuranic-free. Soluble trace quantities of transuranics, well under the ERDAM-0524 concentration guides for uncontrolled areas, will be mixed in the plant effluent and discharged to the Great Miami River."

a. Plutonium Less Than or Equal To 10 Parts Per Billion

Sampling of incoming feeds is accomplished as necessary to assure uranium accountability. Sampling for contaminants is only performed on an exception basis and is performed primarily when a particular

concern is made known to the FMPC staff. Sampling of the in-process material for transuranics is done on a routine basis only at the UO₃ stage in the last few years. It was reported to the Task Force that FMPC products were sampled only sporadically for transuranics and not at all for fission products. FMPC is currently developing a sampling program plan.

During the processing of feed material containing less than or equal to 10 ppb plutonium, the FMPC takes no additional precautions regarding worker protection beyond that required for virgin uranium since FMPC determined that the uranium was the controlling element for worker protection.

Task Force Observation

Existing administrative and radiation protection practices, if correctly applied, would allow the processing of recycle materials containing similar amounts of plutonium. Such practices would include (a) obtaining contaminant data on all incoming feed materials, (b) worker education and training, (c) attention to housekeeping details, (d) improved procedures, and (e) additional sampling and analyses (air, liquid effluents, river water, etc.). Where practicable, engineering controls are preferred over administrative controls. The working conditions that generated the highest plutonium levels should not be repeated.

b. Plutonium Greater Than 10 Parts Per Billion

The Paducah Gaseous Diffusion Plant shipped residual ash (resulting from the processing Hanford returns) from the Paducah Feed Plant (which was shutdown in 1977) to the FMPC on at least two different occasions to recover the uranium in the ash (in 1975-1976 and again in 1980). In June 1980, 22.5 metric tons of uranium (MTU) contained in

about 40 metric tons of ash from the processing of Hanford UO_3 returns at Paducah were received at the FMPC in 16 five-ton hopper containers. According to FMPC, the plutonium levels in the 16 hoppers ranged from a low of 67 ppb per uranium to a high of 7,757 ppb per uranium. It was estimated by the FMPC that 25.29 grams of plutonium were contained in these 16 hoppers. The average plutonium concentration measured by FMPC (1,123 ppb Pu-239) was above the maximum plutonium content of 940 ppb Pu-239 measured at the Paducah Feed Plant. It should be noted that at the FMPC (1) only a limited number of samples were taken, (2) the samples were taken mainly at top surface areas, and (3) the material was non-homogeneous.

The FMPC determined that plutonium would be the controlling nuclide in the material and that special handling measures would be necessary due to its higher plutonium content.

Task Force Observation

It should be noted that this material was shipped to the FMPC with DOE's approval. Although it could not be documented whether DOE was aware of the higher levels of plutonium in the Paducah Feed Plant ash, the FMPC staff stated that DOE was verbally notified. From available records, it was noted that the FMPC was aware of elevated levels of transuranics in the Feed Plant Ash. Additionally, it was established that the FMPC agreed to accept and process this material.

Once the material had been accepted, sampled, and analyzed at the FMPC, five of the 16 hoppers were repackaged into drums in the Green Salt Plant (Plant 4). Since this repackaging required open air transfer and some handwork with metal rods to break-up the solidified material, the workers were requested to wear half-mask airline respirators, and health physics attention was given to air sampling. Forty high volume, general air samples were collected at two locations near the work area; breathing zone (Lapel) samples were taken for two

operators; and a sign(s) was posted at the workplace warning workers that the potential for airborne plutonium existed. The results of these samples indicated that, given conservative assumptions regarding solubility and particle size, the maximum airborne plutonium levels were 15 times the DOE maximum permissible concentrations (MPC) for continuous exposure to workers without respiratory protection for the general air at the workstation, and 3 times the MPC for the breathing zone samples. The FMPC determined that the protection factor (1000 to 1) afforded by the respirators (required for this operation) was sufficient to protect the workers. No surveys for transferrable surface contamination were performed.

Task Force Observation

Recent interest has promoted survey attention at the Plant 4 work site used for the repackaging work. The work site was dedicated to this specific repackaging task and has since been simply abandoned in place. On February 22, 1985, a survey was conducted for transferrable contamination at the Plant 4 Hydrogen Burner Bank No. 12; the results indicate some low level Pu-239 contamination is still present at the work site. FMPC plans to decontaminate and restore this work site during the Plant 4 shutdown (late July or early August). Based on DOE and FMPC Health Physics review of this work (in February 1985), overall contamination control was judged to be marginal in that no surveys for transferrable surface contamination were performed at the time of processing to support the effort.

Management awareness of the special monitoring and respiratory protection measures taken for this material was evident. Questions and concerns were raised by the operators and the nearby millwrights regarding special respiratory protection requirements.

Due to the dust being generated by the handwork required to repackage the material, the remaining hoppers were transferred to the Sampling Plant (Plant 1) where the balance of the material was repackaged. FMPC judged that the dusting could be better controlled in the Sampling Plant, although the Task Force was advised that worker protection methods were the same as in the Green Salt Plant.

Task Force Observation

FMPC management stated that special precautions were taken to control exposures at the Sampling Plant; however, the Task Force was not able to document what precautions were taken nor the personnel exposures experienced in the Sampling Plant.

The operating scheme devised by the FMPC to handle this high plutonium material was based on the need to blend plutonium-free (or nearly so) material with the Paducah ash in order to reduce the plutonium concentration to a level where uranium would be the controlling nuclide, thus requiring only routine worker protection controls. The environmental effects of processing the Paducah ash at the FMPC were informally considered prior to the commencement of operations based on information from Paducah as well as results from FMPC laboratory test programs. The conclusion reached by the FMPC staff was that the processing of the Paducah ash could be conducted in an environmentally acceptable manner.

After the processing program (including the blending operation) was initiated, sufficient quantities of "low plutonium or plutonium-free" uranium were introduced with a portion of the Paducah ash to achieve an overall plutonium dilution of 66 to 1. Metal derbies were produced in this campaign, and these derbies have been shipped to meet customer requirements.

Not all of the inventory of Paducah ash was diluted to the ratio stated above. Some of the ash received only a 16 to 1 dilution. At this ratio, about 168 metric tons of uranium (MTU) as UO_3 were generated. This material remains stored at the FMPC pending further processing. The Task Force was advised that the FMPC desires to use this material as blending stock to meet current DOE requirements. Further processing of this material will require special DOE approval to assure that adequate precautions are taken.

Based on the additional samples and analyses performed to support similar work performed in 1975-1976, the FMPC staff concluded that about 87 percent of the plutonium in the feed material stayed with the UO_3 product while the balance moved to the raffinate stream. Solids in the raffinate stream settled out in the large settling basin (Pit 5). Liquids in the raffinate stream were combined with other FMPC liquid streams (such as sewage plant effluent and storm sewer water) which flowed by gravity in a buried pipeline to the Great Miami River. About 42 percent of the neptunium stayed with the product while 58 percent went with the raffinates. Laboratory work at the FMPC has demonstrated that almost all the transuranic activity in the raffinate precipitates during lime neutralization and appears in the filter cake which is eventually buried in FMPC onsite pits.

5. Environment, Safety and Health (ES&H) Impacts

a. Overall ES&H Impacts

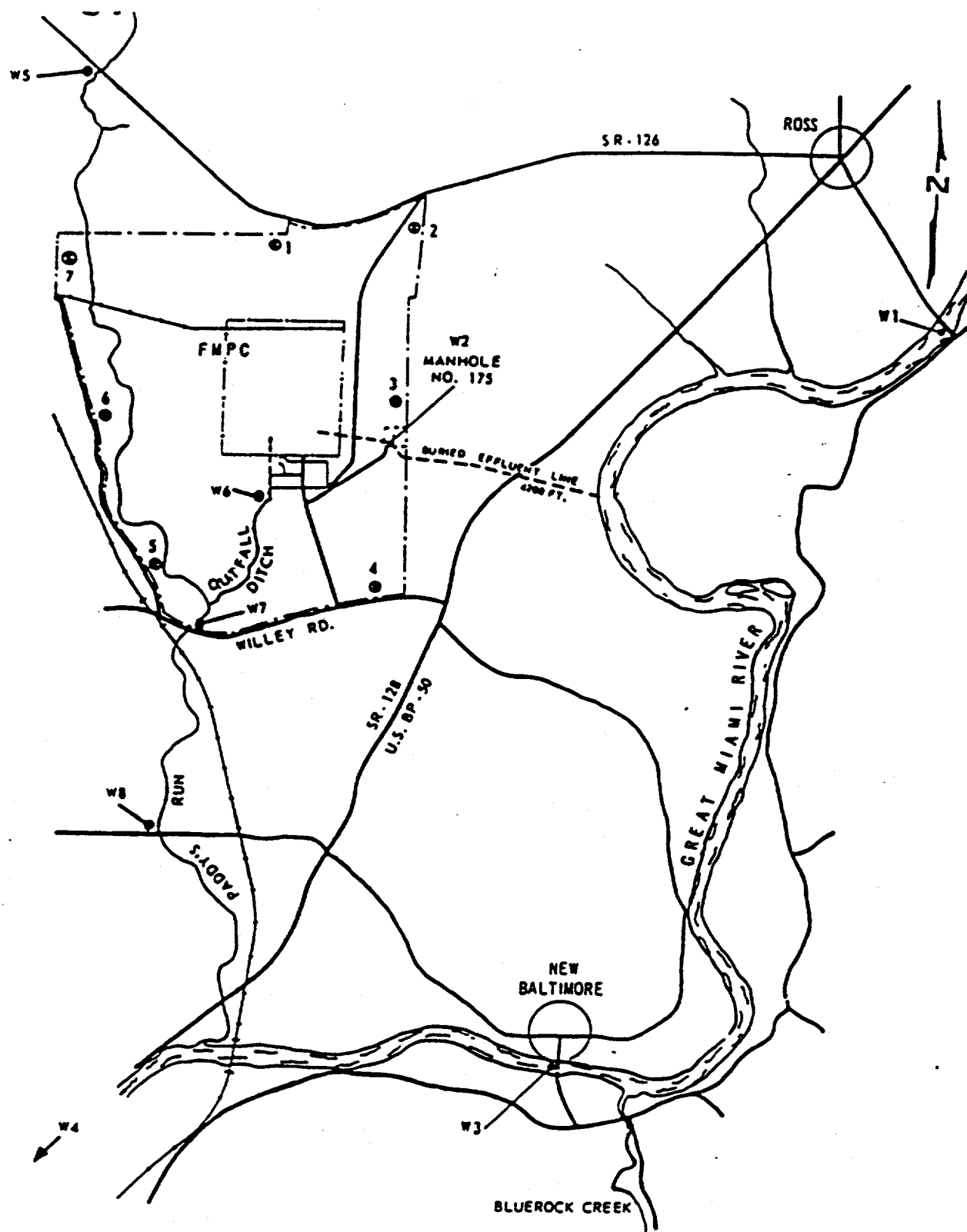
DOE radiation protection criteria for air and water in uncontrolled areas are used as guides for environmental monitoring purposes. Samples collected at the plant boundary are used to determine compliance with criteria established for uncontrolled areas. River samples are collected downstream from the plant effluent discharge point which is upstream from any known use of water as a source of drinking water. DOE criteria for contaminants in water for uncontrolled areas are applied to these samples.

Criteria for nonradioactive materials in air and water are related to state OEPA standards (Ohio Environmental Protection Agency, "Ambient Air Quality Standards" and "Water Quality Standards Administrative Code Chapter 3745-1," effective February 14, 1978). OEPA water quality standards apply beyond the mixing zone allowed for industrial and municipal effluents.

Overall ES&H impacts were considered by the Task Force. The following information contained in this section of the Task Force report was extracted from annual environmental monitoring reports (primarily from the FMPC report number NLC0-2018, dated August 1984, which presents 1983 data). The sampling points noted in Tables 5-11 are based on the sampling locations shown in Figure 4.

Table 5 (from NLC0-2018) shows the air concentration activity for Np-237, Pu-238 and Pu-239 to be small fractions of relevant DOE concentration guides (DOE Order 5480.1A, Chapter XI, Attachment XI-1). For Pu-239 the concentrations for the 7 monitoring stations range from 0.04 to 0.89 percent of the DOE guide for discharge to uncontrolled areas. It is also of interest that the Pu-239 levels in air (for 1983) although small, are a factor of about 10 higher than 1982 levels. The levels given in NLC0-2018 are close to the level reported for fallout Pu-239, 240 (e.g., about 10^{-18} μ Ci/ml air).

In the past most interest in radiation dose from fission products in recycle materials has been directed at radionuclides of Zr-95 and its daughter Nb-95, Ru-106 and its daughter Rh-106 and Ru-103. More recently, interest in Tc-99 has increased. No air concentration data for these particular radionuclides appear in the latest FMPC Environmental Monitoring Annual Report (NLC0-2018). However, data for gross beta concentrations are given. These average air concentration values fall between 0.03 and 0.06 percent of the DOE concentration guide (DOE Order 5480.1A, Attachment XI-1, Table II, note 3) for the seven FMPC site monitoring stations over a 53 week period (Table 6).



W4 is located at Miamitown,
4.7 miles from Paddy's Run.

FIGURE 4 FMPC and Surrounding Area

⊙ BOUNDARY AIR SAMPLING STATIONS.

W1-W8 - WATER SAMPLING LOCATIONS

Table 5
Various Radionuclides in Air - FMPC

Radionuclide	Sampling Point(1)	Concentration Found(2)		Standard(3) μCi/mL
		μCi/mL	% of Standard	
Neptunium-237	BS1	$6.1 \pm 1.3 \times 10^{-17}$	0.061	1×10^{-13}
	BS2	$3.0 \pm 1.3 \times 10^{-17}$	0.030	
	BS3	$8.7 \pm 1.5 \times 10^{-17}$	0.087	
	BS4	$2.3 \pm 0.7 \times 10^{-17}$	0.022	
	BS5	$2.4 \pm 0.7 \times 10^{-17}$	0.024	
	BS6	$4.4 \pm 1.9 \times 10^{-18}$	0.004	
	BS7	$1.3 \pm 0.5 \times 10^{-17}$	0.013	
Plutonium-238	BS1	$1.3 \pm 0.7 \times 10^{-17}$	0.018	7×10^{-14}
	BS2	$1.1 \pm 1.4 \times 10^{-18}$	0.015	
	BS3	$3.2 \pm 1.3 \times 10^{-17}$	0.046	
	BS4	$7.0 \pm 3.5 \times 10^{-18}$	0.010	
	BS5	$6.4 \pm 3.0 \times 10^{-18}$	0.009	
	BS6	$5.4 \pm 2.7 \times 10^{-18}$	0.008	
	BS7	$1.8 \pm 3.5 \times 10^{-19}$	<0.001	
Plutonium-239	BS1	$4.1 \pm 0.9 \times 10^{-16}$	0.683	6×10^{-14}
	BS2	$3.2 \pm 0.9 \times 10^{-16}$	0.533	
	BS3	$5.3 \pm 1.1 \times 10^{-16}$	0.887	
	BS4	$1.7 \pm 0.3 \times 10^{-16}$	0.278	
	BS5	$1.4 \pm 0.2 \times 10^{-16}$	0.240	
	BS6	$9.9 \pm 1.7 \times 10^{-17}$	0.165	
	BS7	$2.3 \pm 0.7 \times 10^{-17}$	0.037	
Thorium-228	BS1	$4.8 \pm 1.1 \times 10^{-17}$	0.024	2×10^{-13}
	BS2	$4.1 \pm 1.6 \times 10^{-17}$	0.021	
	BS3	$9.3 \pm 1.5 \times 10^{-17}$	0.047	
	BS4	$2.6 \pm 0.7 \times 10^{-17}$	0.013	
	BS5	$4.0 \pm 0.9 \times 10^{-17}$	0.020	
	BS6	$2.7 \pm 0.7 \times 10^{-17}$	0.014	
	BS7	$1.4 \pm 0.1 \times 10^{-17}$	0.007	
Thorium-232	BS1	$6.1 \pm 1.3 \times 10^{-17}$	0.006	1×10^{-12}
	BS2	$3.9 \pm 1.6 \times 10^{-17}$	0.004	
	BS3	$1.4 \pm 0.2 \times 10^{-16}$	0.010	
	BS4	$5.1 \pm 1.0 \times 10^{-17}$	0.005	
	BS5	$4.3 \pm 0.9 \times 10^{-17}$	0.004	
	BS6	$1.9 \pm 0.5 \times 10^{-17}$	0.002	

Footnotes:

- (1) See sampling locations shown in Figure 4.
- (2) Concentration of a composite of 53 weekly samples.
- (3) DOE Order 5480.1A, Attachment XI-1, Table II.

Routinely Monitored Radioactive Contaminants in Air - FMPC

Contaminant	Sampling Point (1)	Number of Samples	Maximum Concentration Found		Maximum Concentration Found		Average Concentration			95% Confidence Level	Detection Level $\mu\text{Ci/mL}$	Standard $\mu\text{Ci/mL}$
			$\mu\text{g/m}^3$	$\mu\text{Ci/mL}$	$\mu\text{g/m}^3$	$\mu\text{Ci/mL}$	$\mu\text{g/m}^3$	$\mu\text{Ci/mL}$	% of Standard			
Uranium	BS1	53	0.295	2.0×10^{-13}	0.0009	6.3×10^{-16}	0.031	2.1×10^{-14}	0.5	$+2 \times 10^{-15}$ $-\mu\text{Ci/mL}$	1×10^{-16}	4×10^{-12} (2)
	BS2	53	0.145	9.8×10^{-14}	0.0002	1.6×10^{-16}	0.021	1.4×10^{-14}	0.3			
	BS3	53	0.369	2.5×10^{-13}	0.0004	2.6×10^{-16}	0.036	2.5×10^{-14}	0.6			
	BS4	53	0.130	8.8×10^{-14}	0.0006	3.9×10^{-16}	0.013	8.9×10^{-15}	0.2			
	BS5	53	0.148	1.0×10^{-13}	0.0005	3.5×10^{-16}	0.014	9.8×10^{-15}	0.2			
	BS6	53	0.068	4.6×10^{-14}	0.0011	7.5×10^{-16}	0.017	1.1×10^{-14}	0.3			
	BS7	53	0.055	3.7×10^{-14}	0.0005	3.0×10^{-16}	0.007	4.8×10^{-15}	0.1			
Gross Beta	BS1	53	NA(3)	2.3×10^{-13}	NA	1.0×10^{-14}	NA	5.6×10^{-14}	0.06	$+6 \times 10^{-15}$ $-\mu\text{Ci/mL}$	1×10^{-16}	1×10^{-10} (4)
	BS2	53		1.3×10^{-13}		9.8×10^{-15}		4.1×10^{-14}	0.04			
	BS3	53		2.5×10^{-13}		8.0×10^{-15}		5.7×10^{-14}	0.06			
	BS4	53		1.1×10^{-13}		9.6×10^{-15}		2.8×10^{-14}	0.03			
	BS5	53		9.1×10^{-14}		1.2×10^{-14}		3.1×10^{-14}	0.03			
	BS6	53		8.4×10^{-14}		1.3×10^{-14}		3.7×10^{-14}	0.04			
	BS7	53		3.6×10^{-13}		9.9×10^{-15}		3.2×10^{-14}	0.03			

(1) See sampling locations in Figure 4.

(1) See sampling locations in Figure 4.
(2) DOE Order 5480.1A, Attachment XI-1, Table II Concentration Guide for Natural Uranium based on one gram of natural uranium containing 6.77×10^{-7} Ci/g. Past FPMC Environmental Monitoring Annual Reports defined one curie of natural uranium as equivalent to 3,000 kg of natural uranium (or 3.33×10^{-7} Ci/g) per the guidelines presented in DOE Order 5480.1A, Attachment XI-1, Table II. In this report the definition of a curie of natural uranium will be as defined in 10 CFR 20 (§§ for 6.77×10^{-7} Ci/g).

(3) Not applicable.

(4) DOE Order 5480.1A, Attachment XI-1, Note 3, Concentration Guide for Mixture of Unidentified Radionuclides.

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NLCO-2018 contains certain information on radionuclides discharged from water sampling point W2 at FMPC (the final control point before release to the Great Miami River). The value for Tc-99 is 11 percent of the DOE guide for discharge to uncontrolled areas. It is important to note, however, that the standard or guide is not for drinking water that may be used by human beings. This guide, therefore, should not be applied to the plant effluent at station W2. (Dose standard compliance at the FMPC is determined after mixing of effluents in the Great Miami River -- not at the release point.) Tc-99 is the third most prevalent radioactive element in the wastewater discharge based upon comparison to concentration guides and the largest contributor from a total curie standpoint. These data are shown in Table 7.

Table 8 gives information on the concentration of Uranium, Ra-226, Ra-228, gross alpha and gross beta in water samples for 1983. Sampling stations W7 and W8 are consistently elevated for uranium, gross alpha and gross beta. The gross alpha and gross beta measurements represent significant percentages of the DOE concentration guide for uncontrolled areas (up to 50 percent of the gross beta [less background] and 26 percent for gross alpha at station W8.) Public exposure from this discharge before mixing in the Great Miami River is not expected.

NLCO-2018 does acknowledge that technetium is present in trace quantities in various materials sent to FMPC for uranium recovery, and that analysis for Tc-99 in sediments was initiated in 1983. Sediments obtained within the FMPC site and offsite do contain trace quantities of Tc-99 (Tables 9 and 10).

b. Routine Processing of Recycled Uranium

Respiratory protection requirements were addressed by the Task Force. Section 7 of the NLO, Inc. Health and Safety Manual contains information on the respiratory protection program. Section 7.3.3

Table 7

Radionuclides Discharged Via Sampling Point W2(1) - FMPC

Radionuclide	Total Curies	Average Concentration Found)		Standard(2) $\mu\text{Ci/mL}$
		$\mu\text{Ci/mL}$	% of Standard	
Cesium-137	5.6×10^{-3}	8.6×10^{-9}	0.04	2×10^{-5}
Neptunium-237	$<1.8 \times 10^{-4}$	$<2.8 \times 10^{-10}$	<0.009	3×10^{-6}
Plutonium-238	5.1×10^{-6}	7.8×10^{-12}	<0.001	5×10^{-6}
Radium-226	1.4×10^{-3}	2.2×10^{-9}	7.3	3×10^{-6}
Radium-228	6.2×10^{-3}	9.5×10^{-9}	32	3×10^{-6}
Ruthenium-106	3.1×10^{-4}	4.8×10^{-10}	0.005	1×10^{-5}
Strontium-90	5.9×10^{-3}	9.1×10^{-9}	3	3×10^{-7}
Technetium-99	2.1×10^1	3.2×10^{-5}	11	3×10^{-4}
Thorium	2.3×10^{-4}	3.5×10^{-10}	0.04	1×10^{-6}
Uranium (3)	4.0×10^{-1}	6.1×10^{-7}	51	1.2×10^{-6}

Footnotes:

- (1) Radionuclides in the plant effluent which is discharged to the Great Miami River through a buried pipeline. An additional 3.0×10^{-2} Curie of uranium was contained in the storm sewer overflow discharged into a ditch at sampling point W6. The ditch empties into Paddy's Run above sampling point W7.
- (2) DOE Order 5480.1A, Attachment XI-1, Table II, Concentration Guides for Water in Uncontrolled Areas. These Guides are for water such as the Great Miami River and are not meant to be applied to the plant effluent. They are listed here for comparison purposes.
- (3) Curies of natural uranium using the 10 CFR 20 definition of natural uranium activity.

Table 8

Radioactive Contaminants in Water - FAPC

Contaminant	Sampling Point(1)	Number of Samples(2)	Maximum Concentration Found		Maximum Concentration Found		Average Concentration			95% Confidence Limits(3)	Detection Level $\mu\text{Ci/mL}$	Standard $\mu\text{Ci/mL}$
			$\mu\text{g/mL}$	$\mu\text{Ci/mL}$	$\mu\text{g/mL}$	$\mu\text{Ci/mL}$	$\mu\text{g/mL}$	$\mu\text{Ci/mL}$	% of Standard			
Uranium	W1	52	0.005	3.4×10^{-9}	0.001	6.7×10^{-10}	0.002	1.4×10^{-9}	0.12	$+1.4 \times 10^{-9}$ — $\mu\text{Ci/mL}$	6.7×10^{-10}	1.2×10^{-6} (5)
	W3	52	0.006	4.1×10^{-9}	0.001	6.7×10^{-10}	0.003	2.0×10^{-9}	0.17			
	W4	52	0.005	3.4×10^{-9}	0.001	6.7×10^{-10}	0.003	2.0×10^{-9}	0.17			
	W5	49	0.005	3.4×10^{-9}	<0.001	6.7×10^{-10}	0.002	1.4×10^{-9}	0.12			
	W7	26	0.908	6.1×10^{-7}	0.003	2.0×10^{-9}	0.112	7.6×10^{-8}	6.3			
	W8	26	0.044	3.0×10^{-8}	0.006	4.1×10^{-9}	0.013	8.8×10^{-9}	0.73			
Radium-226	W1	12	NA(4)	4.5×10^{-10}	NA	4.5×10^{-10}	NA	4.5×10^{-10}	<1.5	$+4.5 \times 10^{-10}$ — $\mu\text{Ci/mL}$	4.5×10^{-10}	3×10^{-8} (5)
	W3	12		4.5×10^{-10}		4.5×10^{-10}		4.5×10^{-10}	<1.5			
	W4	12		4.5×10^{-10}		4.5×10^{-10}		4.5×10^{-10}	<1.5			
	W5	6		9.0×10^{-10}		4.5×10^{-10}		4.5×10^{-10}	<1.5			
	W7	10		4.5×10^{-10}		4.5×10^{-10}		4.5×10^{-10}	<1.5			
	W1	12		9.0×10^{-10}		4.5×10^{-10}		4.5×10^{-10}	<1.5			
Radium-228	W3	12	NA	1.4×10^{-9}	NA	4.5×10^{-10}	NA	4.5×10^{-10}	<1.5	$+4.5 \times 10^{-10}$ — $\mu\text{Ci/mL}$	4.5×10^{-10}	3×10^{-8} (5)
	W4	12		9.0×10^{-10}		4.5×10^{-10}		4.5×10^{-10}	<1.5			
	W5	6		1.4×10^{-9}		4.5×10^{-10}		9.0×10^{-10}	3.0			
	W7	10		1.8×10^{-9}		4.5×10^{-10}		9.0×10^{-10}	<3.0			
	W1	12		4.5×10^{-9}		9.0×10^{-10}		4.5×10^{-9}	<7.7			
	W3	52		4.5×10^{-9}		9.0×10^{-10}		4.5×10^{-9}	<7.7			
Gross Alpha	W4	52	NA	6.3×10^{-9}	NA	9.0×10^{-10}	NA	4.5×10^{-9}	<7.7	$+8.0 \times 10^{-10}$ — $\mu\text{Ci/mL}$	9.0×10^{-10}	3×10^{-8} (6)
	W5	50		5.9×10^{-9}		9.0×10^{-10}		4.5×10^{-9}	<7.7			
	W7	26		5.8×10^{-7}		2.3×10^{-9}		5.0×10^{-9} (7)	17(7)			
	W8	26		2.3×10^{-8}		4.1×10^{-9}		7.7×10^{-9}	26			
	W1	52		1.2×10^{-8}		2.7×10^{-9}		5.9×10^{-9}	20			
	W3	52		5.1×10^{-8}		3.2×10^{-9}		9.5×10^{-9}	32			
Gross Beta	W4	52	NA	3.4×10^{-8}	NA	3.6×10^{-9}	NA	8.6×10^{-9}	29	$+3.0 \times 10^{-9}$ — $\mu\text{Ci/mL}$	2.7×10^{-9}	3×10^{-8} (6)
	W5	50		3.2×10^{-8}		2.7×10^{-9}		6.3×10^{-9}	21			
	W7	26		1.3×10^{-7}		3.2×10^{-9}		1.6×10^{-8}	53			
	W8	26		8.7×10^{-8}		2.7×10^{-9}		2.1×10^{-8}	70			
	W1	52		1.2×10^{-8}		2.7×10^{-9}		5.9×10^{-9}	20			
	W3	52		5.1×10^{-8}		3.2×10^{-9}		9.5×10^{-9}	32			

Footnotes:

- (1) See sampling locations shown in Figure 4.
- (2) Samples are composited for radium analyses as follows: one-month composites of daily samples from W1 and W3; one-month composites of weekly samples from W4, and all available weekly samples from W7; two-month composites of weekly samples from W5.
- (3) Confidence Limits are given for the average concentration.
- (4) Not applicable.
- (5) DOE Order 5480.1A, Attachment XI-1, Table 11, Concentration Guides for natural uranium, radium-226 and radium-228.
- (6) DOE Order 5480.1A, Attachment XI-1, Note 2, Concentration Guides for mixtures of unidentified radionuclides.
- (7) Since the standard is for unidentified alpha activity, this figure is based on the difference between total gross alpha activity and uranium alpha activity.

Table 9
Radionuclides in FMPC Onsite Sediment

Sampling Point (2)	Uranium Concentration (1)			Technetium-99 Concentration		
	$\mu\text{Ci/g}$	95% Confidence Level	Detection Level	$\mu\text{Ci/g}$	95% Confidence Level	Detection Level
1	1.7×10^{-6}	<u>+25%</u>	3.4×10^{-7} $\mu\text{Ci/g}$	$<1.4 \times 10^{-7}$	<u>+20%</u>	1.4×10^{-7} $\mu\text{Ci/g}$
2	3.2×10^{-5}					
3	1.8×10^{-4}					
4	1.3×10^{-4}			8.1×10^{-6}		
5	1.5×10^{-6}			1.4×10^{-6}		
6	1.9×10^{-4}			1.7×10^{-5}		
7	6.9×10^{-5}					

Footnotes:

- (1) Results on dry basis.
(2) See sediment sampling locations shown in Figure 4.

Table 10

Radionuclides in Great Miami River Sediment - FMPC

Sampling Point (2)	Distance From FMPC Outfall	Uranium Concentration (1)			Technetium-99 Concentration		
		$\mu\text{Ci/g}$	95% Confidence Level	Detection Level	$\mu\text{Ci/g}$	95% Confidence Level	Detection Level
8 9	Upstream						
	3.7 miles	1.8×10^{-6}	$\pm 25\%$	3.4×10^{-7} $\mu\text{Ci/g}$	$<1.4 \times 10^{-7}$	$\pm 20\%$	1.4×10^{-7} $\mu\text{Ci/g}$
	1.5 miles	1.7×10^{-6}					
10 11 12 13 14	Downstream						
	50 feet	3.1×10^{-6}	$\pm 25\%$	3.4×10^{-7} $\mu\text{Ci/g}$	4.2×10^{-6}	$\pm 20\%$	1.4×10^{-7} $\mu\text{Ci/g}$
	0.8 mile	1.3×10^{-6}			9.0×10^{-7}		
	3.3 miles	1.6×10^{-6}					
	4.5 miles	2.7×10^{-6}			1.1×10^{-6}		
	4.7 miles	2.1×10^{-6}					

Footnotes:

- (1) Results on dry basis.
 (2) See sediment sampling locations shown in Figure 4.

(Respiratory Protective Equipment Requirements) states that air-supplied hood or air-line respirators are required for work at locations in which the National Lead Company of Ohio Concentration Guide (NCG) for airborne radioactivity could be exceeded by a considerable margin but the condition is not immediately dangerous to life or health. The NCG value is 100 dpm (alpha) which is about 45 percent of the U.S. NRC Concentration Guide of 220 dpm for alpha emitting radionuclides.

From an environmental standpoint, environmental measurements for plutonium and neptunium were not made until 1982. The values given in the FMPC Environmental Monitoring Annual Reports for 1982 (May 1983, NLC0-1187) and 1983 (August 1984, NLC0-2018) for airborne plutonium (Pu-239 and Pu-238) and concentrations of Np-237 at selected monitoring locations around the plant boundary are for a composite of 53 weekly samples. The FMPC radiometric procedure for plutonium (A-03-0279) is dated June 28, 1976 (revised, February 1979).

Task Force Observation

The plutonium radiometric procedure is not state-of-the-art. For example, a Np-237 spike is used in the procedure. Most analyses currently use Pu-242 or Pu-236 for this purpose which provides increased precision. Also, for low concentrations of plutonium one should use chemical separation of the plutonium followed by electro-deposition and alpha spectrometry. A state-of-the-art procedure is available from the Los Alamos National Laboratory (Manual of Analytical Methods for Radiobioassay, LA-9763-M, LANL, July 1983; compiled by M. A. Gautier). The Task Force has offered a recommendation to update FMPC's analytical and measurement capabilities to state-of-the-art levels. It is not clear when measurements for transuranic radionuclides were initiated for recycle materials or plant products. There is little FMPC data on transuranics in FMPC products.

c. Paducah Feed Plant Ash Campaign

It was reported by the FMPC that some of the uranium concentrations measured for the repackaging of the Paducah Feed Plant ash were up to 100 times above the NCG values. Thirty-two percent of the samples exceeded the NCG. For the plutonium measurements, it was conservatively assumed that all the plutonium was soluble in form, thus a DOE guide of 2×10^{-12} $\mu\text{Ci/ml}$ or 4.4 alpha dpm per m^3 was used. At the hopper station, plutonium concentrations ranged from 0.1 to 65 alpha dpm per m^3 ; at the drumming station, values ranged from 0.03 to 49 alpha dpm per m^3 . The DOE guide was exceeded by 52 percent of the samples. None of the plutonium concentrations, however, exceeded the DOE guide for insoluble plutonium which respirable fines in air dispersal are practically certain to be.

Task Force Observation

FMPC management has identified a number of program improvements for control of worker exposure to transuranium radionuclides. One such improvement is to obtain contaminant data on all incoming materials. To further support this effort, shippers should also determine the level of contaminants and certify these levels for FMPC. As a double check, measurements should also be made at the FMPC.

The Task Force was advised that the FMPC plans to purchase a whole body "in-vivo" counter. The in-vivo counter (proposed for FY 1985) should be pursued, and expert advice should be obtained on this acquisition. To the extent possible, the measurement of plutonium in the body should be an objective of the in vivo counter. Plutonium chest burdens probably cannot be accurately measured to levels below about 20 nanocuries Pu-239 using the best of existing equipment, including a well shielded counting facility. Uranium in the lung may also complicate measurement of plutonium. A urine bioassay might be a

better investment of resources and staff, and LANL should be contacted for assistance. In early July 1985, several FMPC workers that participated in the 1982 processing campaign of the Paducah ash were sent to Richland, Washington for plutonium burden measurements. The results of these workers were all negative as to plutonium uptake.

Worker protection could have been much better. For example, workers should have been better informed. The NLO-FMPC Manufacturing Standards/Procedures (November 17, 1982, revised January 7, 1985) mention only briefly the need for air-purifying respirator with purple radionuclide cartridges when dusty conditions exist. The standard also appears to place the burden of deciding whether or not a significant uptake of radionuclides has occurred (and thus the need for urine analysis) on the employee.

Overall, additional environment, safety and health management attention and involvement is required. There is a need for better respiratory protection, housekeeping practices, and radiation protection and control practices at FMPC. The need for operator education, especially when working with materials in excess of 10 ppb plutonium, exists as does the need for meaningful operator training.

The need exists for management at FMPC to thoroughly review the manner in which the Paducah Feed Plant ash has been processed and prepare a detailed procedure to address handling of recycle materials that may be high (greater than 10 ppb Pu-239) in transuranic concentrations should the occasion arise again. Even the uranium concentrations were above the normal operating conditions during this campaign.

During the preparation of this report, the Task Force became aware of three special environment, safety and health reviews of the FMPC. These included:

1. *Health Physics Review of the FMPC by F. C. Gilbert, et. al.*
2. *Nuclear Safety Program Appraisal of the Oak Ridge Operations Office by the DOE-Headquarter Office of Policy, Safety and Environment.*
3. *Environmental Program Review of the FMPC by the Oak Ridge Associated Universities (draft report only).*

The Task Force reviewed these special reports and concurred in the pertinent findings and recommendations that were applicable or overlapped. The FMPC Health Physics and Environmental Program Reviews are especially noteworthy and were, for the most part, confirmed by the Task Force during the onsite review. The Task Force decided that, rather than repeating findings and recommendations, it would be more prudent to endorse those reports and encourage the FMPC staff to vigorously implement those recommendations already offered.

The more salient points from each review are briefly discussed below.

- o Health Physics Review: (1) since the interior of operating plant buildings were dirty, a thorough cleaning was needed to reduce the potential for unnecessary radiation exposure; (2) the technical depth of the health physics staff was insufficient to maintain the program at an adequate level and to accomplish required improvements; (3) radiation training/retraining for workers was not sufficiently documented to verify adequate and required training had been accomplished; (4) required dosimetry records were available but required extensive search to gather individual data; (5) deficiencies in some emergency equipment were noted; and (6) even though the FMPC Health Physics program had many shortcomings, no evidence was found that the employees' health and safety were in jeopardy.*

- o Nuclear Safety Program Appraisal: (1) fixed continuous air samplers did not exist in FMPC facilities, but rather spot breathing zone samples are taken where warranted; air sampling records indicated that workers in certain operations have regularly exceeded airborne contamination limits for uranium (MPCs), and prescribed actions are neither documented nor implemented; (2) FMPC does not have contamination survey instruments at the work site for use in checking for skin and clothing contamination; it was questionable whether appropriate Health Physics procedures had been applied in the processing of transuranic contaminated material; (3) the routine surveillance program to monitor internal exposure of workers consisted of both in-vivo and excreta measurement, but there was little evidence to suggest that any dose assessments were being performed.
- o Environmental Program Review: (1) several shortcomings were identified by this review team, however, it was noted that the FMPC had already taken measures to correct many of the deficiencies; (2) deficiencies were identified in the areas of (a) organizational structure - the environmental program was structurally fragmented, (b) data analysis - there appeared to be no direction in the data analysis effort, (c) terrestrial and sediment sampling - there appeared to be a general absence of coordination in the sampling design and plant activities, (d) air monitoring - air monitors were at best measuring fugitive emissions and plume downwash contributions, (e) water and wastewater monitoring - additional monitoring was required, (f) groundwater - there was a lack of ability to properly characterize the existing groundwater system at the FMPC, (g) waste management - FMPC waste management activities had resulted in contaminant release to the environment, (h) laboratory procedures - on the whole, the analytical laboratory procedures were very good, (i) quality assurance - no formal quality assurance program was in place for the ambient air monitoring program, (j) engineering information - a major deficiency at FMPC was the lack of complete engineering information on the varied sources.

B. The Y-12 Plant

1. Description of Physical Plant

a. General Overview

The Y-12 Plant is a large, multipurpose uranium manufacturing facility which utilizes an extensive technological base and sophisticated fabrication capabilities in support of a variety of DOE programs. The main mission of the Y-12 Plant is the production of materials and components for nuclear weapons.

Although many operations support the Y-12 Plant mission, further discussion on the operations at the Y-12 Plant will be limited to those uranium processing areas directly concerned with the flow of enriched uranium recycle material from the Savannah River Plant (SRP) and the Idaho Chemical Processing Plant (ICPP) to Y-12.

b. Enriched Uranium Recovery and Processing

The economic value of enriched uranium requires the reclamation of most uranium side streams generated during fabrication and production processes. Enriched uranium generated by DOE installations (such as the SRP and ICPP) provides a supply of materials for recovery and processing.

Recovery operations consist basically of (1) burning combustibles, (2) dissolving or leaching solids, and (3) purifying the uranium bearing solutions by liquid-liquid extraction. The purified uranyl nitrate hexahydrate (UNH) solutions are denitrated to uranium trioxide (UO_3) which is then reduced with hydrogen to uranium dioxide (UO_2). This UO_2 is subsequently converted to uranium tetrafluoride (UF_4) by reaction with gaseous anhydrous hydrogen fluoride. Finally, the UF_4

from this process is reduced under high temperatures to yield uranium metal which can then be cast into desired mold configurations. At almost any point in this operation, the enriched uranium can be withdrawn to meet a particular material need. Additionally, various enriched uranium streams can be introduced into the process by matching the various material forms. (See Figure 5).

2. Sources of Enriched Uranium

The two material flows coming into the Y-12 Plant that are addressed by this report are those originating from the SRP and ICPP. Both flows are processed as described above with the point of introduction into the Y-12 operation being the only significant difference.

The enriched uranium from the SRP is received at the Y-12 Plant in Department of Transportation approved shipping containers (tanker trailers usually containing 3,800 - 5,000 gallons) in the form of UNH solution. After unloading and sampling, the SRP material is evaporated to a concentration level that is more amenable to further processing. The enriched uranium from the ICPP is received as UO_3 . After unloading and sampling, the ICPP material is dissolved in nitric acid to yield a UNH solution amenable to further processing. The processing of these two flows is shown in Figure 5.

At Y-12, the UNH is purified by solvent extraction, processed to metal, and returned to the SRP for fabrication into new fuel elements. The enriched uranium received from the ICPP is processed in a manner similar to that from the SRP and is eventually returned to the SRP for further use.

The Y-12 Plant began receiving enriched uranium for processing as described above in fiscal year 1953 from the ICPP and in fiscal year 1955 from the SRP. Estimates of total recycle material receipts from these sources are given in Table 11 below.

MATERIAL PROCESS FLOW

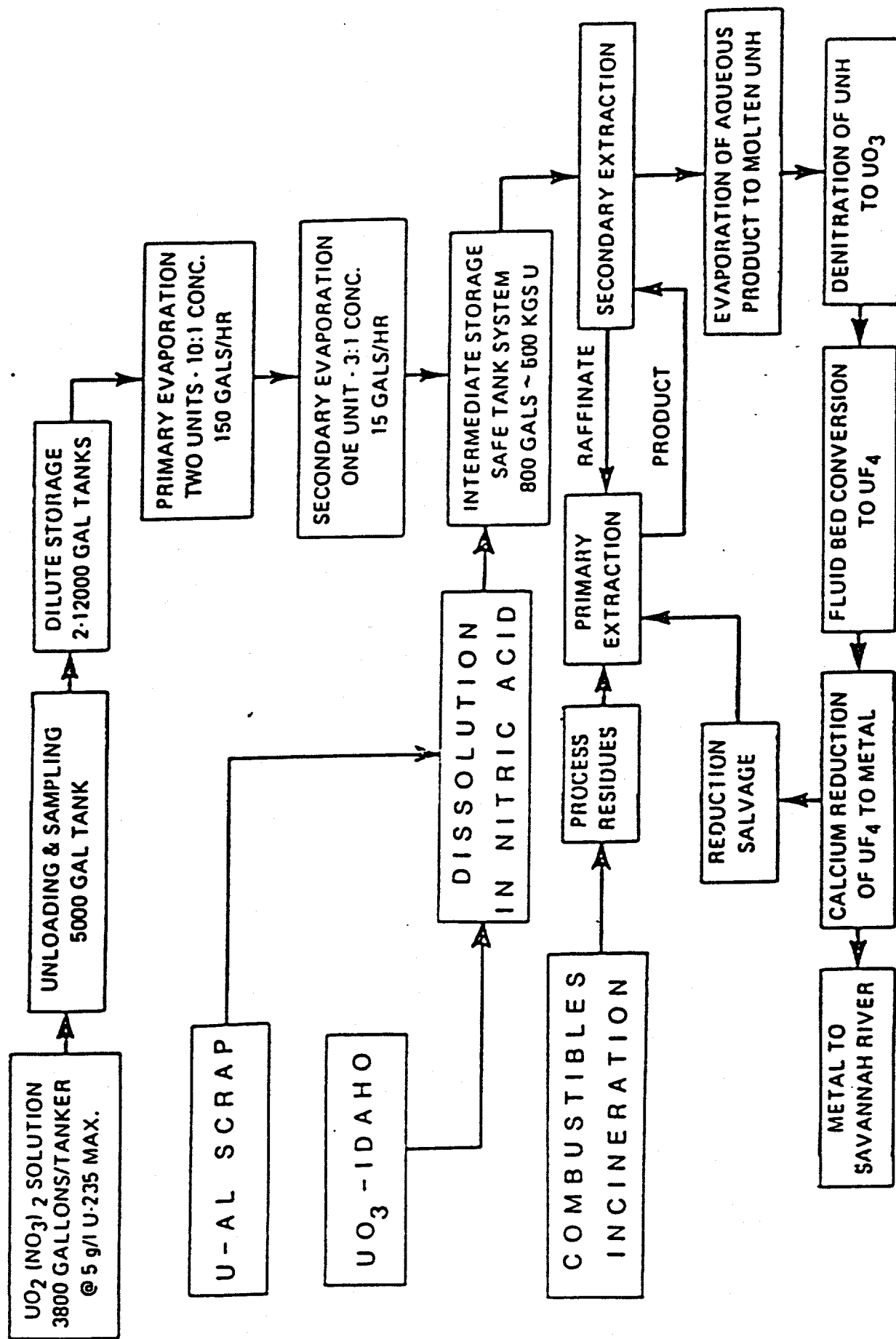
Figure 5
-42-

Table 11

Y-12 Estimated Receipts of Recycle Materials (Kg. U)

<u>Fiscal Year</u>	<u>SRP</u>	<u>ICPP</u>	<u>Total</u>
1953	0	101	101
1954	0	217	217
1955	3	828	831
1956	0	744	744
1957	201	797	998
1958	258	898	1,156
1959	270	3,741	4,011
1960	6,395	769	7,164
1961	2,305	0	2,305
1962	2,701	775	3,476
1963	6,461	0	6,461
1964	2,977	771	3,748
1965	3,546	425	3,971
1966	3,467	1,408	4,875
1967	2,604	0	2,604
1968	2,097	394	2,491
1969	4,121	427	4,548
1970	2,045	108	2,153
1971	3,805	1,660	5,465
1972	4,716	415	5,131
1973	5,051	563	5,614
1974	4,599	0	4,599
1975	5,110	1,702	6,812
1976*	4,320	195	4,515
1977	4,497	1,333	5,830
1978	2,070	525	2,595
1979	4,591	535	5,126
1980	1,510	0	1,510
1981	4,918	905	5,823
1982	5,728	577	6,305
1983	6,682	1,041	7,723
1984	5,776	2,868	8,644
Totals (FY 1953-1984)	102,824	24,722	127,546

*Includes the 3-month transition quarter.

3. Standards/Specifications

At the present time, formal specifications on the maximum quantities of transuranics and fission product elements do not exist between the Y-12 Plant and the feed material suppliers (SRP and ICPP). Y-12 reported that such specifications were suggested by the Y-12 staff at one time; however, the working level staffs were able to mutually resolve any concerns at that time and, as a result, formal specifications were never adopted. Informal specifications in the form of "gentlemen's agreements" did evolve and have been in use since.

Task Force Observation

The Task Force observed that there were some differences in the informal specifications currently in use at Y-12 and the SRP. Formal, working specifications are preferred over informal (gentlemen's agreements) specifications.

These informal specifications used by Y-12 are based on the activities of radioactive emitters. The specifications and the basis for the specifications are given below:

Alpha Activity

$$\text{Specification} = \text{Alpha Ratio} = \frac{(\text{Activity per Gram of Pu} + \text{Np} + \text{Th}) \times 700}{\text{Nominal Activity of Enriched Uranium}} \leq 1.0$$

Basis: Derived to maintain the relative hazard potential of alpha emitters other than uranium to less than 10 percent, i.e., 7 percent of the relative hazard potential of uranium.

$$\frac{50}{700} (\text{relative hazard of Pu to U}) = 7 \text{ percent}$$

Task Force Observation

Y-12's alpha ratio specification was based on a total alpha specification of about 200,000 dpm alpha/g U.

Beta Activity

$$\text{Specification} = \text{Beta Ratio} = \frac{\text{Activity of Sample}}{\text{Activity of Unirradiated Uranium Standard}} \leq 1.25$$

Basis: Selected arbitrarily as usually being achievable and having little additional exposure potential.

Gamma Activity

$$\text{Specification} = \text{Total Fission Product} \leq 0.20 \mu\text{Ci/g Uranium}$$

Basis: Y-12 personnel stated that an informal agreement existed between the SRP and Y-12 to control fission products. This specification was chosen as being usually achievable and not adding unduly to personnel exposure potential.

4. Processing of Enriched Uranium

The processing of the SRP and ICPP recycle materials is conducted in Buildings 9212 and 9206. Several sampling points are located throughout the processing operation at Y-12. However, samples are taken primarily for uranium accountability, operational safety, and operational control. In general, accountability samples are not taken for transuranic (plutonium and neptunium) or fission product elements introduced, processed, or removed from the process.

In summary form, the current Y-12 recycle material sampling policy associated with transuranic and fission product elements is as follows:

- o Sample each batch of receipts.
- o If batches received are small, samples from batches are composited.
- o Sample one out of every ten of the product batches being returned to the SRP.
- o Sample process side streams on a limited frequency (usually annually).

From an analytical standpoint, each recycle material sample is analyzed for its alpha, beta, and gamma activity as well as for the total gamma. Note that these are not accountability samples. Table 12 outlines the total spectra of analyses currently being performed by Y-12 on recycle material samples.

Based on the results of 66 samples taken during the 1950s from the ICPP the amount of plutonium (as a percent of present specifications) averaged less than the informal Y-12 specification (the highest was 830 percent). However, on the average, the beta activity ratio consistently exceeded the specification. Early SRP (1964-1972) returns, based on 144 samples, generally complied with the informal specification on the average although ten (10) samples did exceed the specification (the highest was 180 percent). Sample results over the most recent eight (8) year period (spanning 214 samples) indicate that 22 samples exceeded the informal specification (the highest was 165 percent); however, on the average, the returns did not exceed the specification. It should be noted that the SRP does not analyze for beta activity or recognize a beta specification.

Task Force Observation

Although most incoming material is accompanied by transuranic data from the generating site, the Task Force noted that batches are not held for Y-12 sample results. Thus, a campaign could be in process based on

Table 12

Y-12 ANALYSES PRESENTLY PERFORMED ON
RECYCLE MATERIAL SAMPLES

Alpha Activity

Pu-238, 239-40
Np-237
Th-228
U-232
Total U Alpha

Beta Activity

$$\text{Beta Ratio} = \frac{\text{Activity of Sample}}{\text{Activity of Uranium Standard}}$$

Gamma Activity, Fission Products

Cs-137
Zr-Nb-95
Ru-106

$$\text{Total Gamma} = \frac{\text{Microgram Ra-226 Equivalent}}{\text{Gram U}}$$

shipper's data when Y-12 sample results become available. Since, historically, receipts have generally averaged less than the Y-12 specifications, the Task Force does not consider this to be an unacceptable practice.

As for the product produced by Y-12, results from the eight most recent years (186 samples) show that there were only five (5) occasions in which the product exceeded the specification.

5. Environment, Safety and Health Impacts

The Task Force examined receipts and shipments for contaminant levels and internal Y-12 process generations to determine if contaminants were concentrating in the process. Based on side stream sampling conducted by Y-12 since about 1977, it was noted that there has been a buildup of contaminants in both the liquid and solid waste streams as a result of recycle material processing. Based on limited data (30 samples), it appears that fission product element concentration in the discards of the waste stream (which previously went to the S-3 Ponds) have increased while concentrations in the product streams have decreased. Recent (November 1984) sampling of the S-3 Pond sludge found Pu-238, Pu-239, Pu-240 and Np-237 in the sludge. A closure plan for the S-3 Ponds has been developed by the Y-12 Plant in conjunction with local, state, and federal agencies.

The exposure records of recycle material workers were compared with those of other workers within the same operating departments. The results of this comparison shows that recycle material workers, on the average, had about 2.7-3.0 times the external radiation exposure as the other workers in the department. In a similar manner and based on employee exposure records, Y-12 has calculated the external exposure of workers handling recycle materials to be about 1.2-1.6 times the exposure of Y-12 workers handling unirradiated material (see Table 13).

Table 13

EXTERNAL EXPOSURE OF Y-12 WORKERS HANDLING RECYCLE MATERIALS

<u>Group</u>	<u>Average Exposure (rem/yr)</u>	
	<u>Skin</u>	<u>Penetrating</u>
Uranium Recycle Workers	0.528	0.305
Unirradiated Uranium Workers	<u>0.430</u>	<u>0.190</u>
Difference	0.098	0.115

This difference is considered to be an estimate of external exposure due to processing reactor returns. Exposures at this level were not considered a significant health risk by Y-12 and the Task Force.

The estimated internal dose to those workers handling recycle materials has been calculated by Y-12 to be about 0.019 rem (committed dose to the bone) per employee. The bases for the calculation are given in Table 14. All exposure calculations are small fractions of the guides given in DOE Order 5480.1A, Chapter XI, Attachment XI-1, "Concentrations in Air and Water Above Natural Background").

Task Force Observation

It is noted that the processing operations at Y-12 reduce the fission product concentration in the product stream while increasing the concentration in the waste streams. Furthermore, estimates of external and internal doses to those plant workers processing recycle materials show these doses to be well within allowable guides. As a result, no adverse health effects are expected at Y-12 from processing recycle materials at the present informal specification level.

Table 14

ESTIMATED INTERNAL DOSE TO Y-12 WORKERS HANDLING RECYCLE MATERIALS

- * Average results from general air samples (60,000) taken in Y-12 areas handling SRP material from 1977 to 1985 were 3 percent of the uranium radioactivity concentration standard. Workers were assumed to be exposed to this level of air contamination.
- * The average alpha ratio for SRP receipts for these years was 30 percent of the specification.
- * The specification is set to control the exposure from plutonium to 7 percent of that from uranium.

Combine these factors:

<u>Uranium Concentration</u>		<u>Alpha Ratio</u>		<u>Pu/U Dose</u>		<u>Plutonium Concentration</u>
Standard		Specification		Specification Level		Standard
0.03	X	0.30	X	0.07	=	0.00063

Convert to dose to bone per year as follows:

<u>Plutonium Concentration</u>		<u>Committed Dose to Bone per Year at Standard Level</u>		<u>Committed Dose (rem)</u>
Standard	X	30 rem	=	0.019
0.00063				

An average committed dose of 0.019 rem/year to the bone per employee was considered an acceptable health risk by the Y-12 staff.

C. Other DOE Sites

1. Savannah River Plant (SRP)

a. Plant Description

SRP's primary function is the production of plutonium, tritium, and other special nuclear materials for the national defense, and for other governmental uses and some civilian purposes.

Current operating facilities include three production reactors, a fuel and target fabrication plant, two chemical separations plants, the Savannah River Laboratory (SRL) which is primarily a process development laboratory to support production operations, and a number of production support facilities. Core and target elements discharged from the production reactors are processed in the chemical separations area at the SRP. Pu-239, Pu-238, enriched uranium and other desirable reactor products are separated from each other and from fission products.

b. Chemical Processing of Recycle Uranium

For highly enriched runs (the normal SRP mode of operation), the enriched uranium fuel tubes are processed in the 221-H facility (canyon). This canyon building is a totally enclosed concrete building 880 ft. long by 125 ft. wide by 75 ft. high. The bottom third of the building is below ground level. Solutions containing high concentrations of fission products with consequent high radiation are treated totally remotely in the hot canyon. The less radioactive solutions are handled in the warm canyon which is less heavily shielded.

The chemical process used is a modification of the purex process (H area modified or HM) in which most of the small quantity of plutonium present is removed as waste in the first cycle. This aqueous waste is stored in 1.1 million gallon steel underground waste tanks. The products recovered are the enriched uranium and Np-237.

Shipments of UNH solution to the Y-12 Plant (the normal operational mode) are made in tank-trailer trucks. The SRP averages 1-4 tank-trailers per week shipped to the Y-12 Plant during HM operations. Solutions of uranium approved for shipment must meet specifications on gamma and alpha activities and fission products content shown in Table 16.

There have been five low enriched (less than 10 percent enrichment) campaigns run in H canyon (all since 1974). For these campaigns, the canyon is operated using a standard purex process. The recovered uranium is treated the same as in the HM process, but the low enriched solutions are sent directly to the FMPC as UNH. There are no more low enriched campaigns in the foreseeable future.

c. Standards/Specifications

Specifications for the highly enriched HM solution are in Table 15. A HM Data Sheet for Product/Shipment providing analytical data on each shipment is sent to Y-12 with the shipment.

Task Force Observation

It should be noted that these specifications, used by the SRP, differ in some respects from those reportedly used by Y-12 for receipt of this same material. There is insufficient coordination between the SRP and Y-12 sites on required specifications.

0...

2. Isotopic Concentration

The isotopic concentration of the uranium supplied to SRP is dependent upon receipts by Y-12. Blending uranium from other sites with SRP uranium shall be approved in advance by SRP.

3. Radioactivity

These specifications apply to the average of any four consecutive shipments.

3.1 SRP Uranyl Nitrate Feed

Control of radioactivity in the recycle uranium metal is dependent upon the radioactivity of the uranyl nitrate feed solutions. SRP uranyl nitrate feed solutions shall meet the specifications in sections D,3.1.1 and D,3.1.2.

3.1.1 Gamma Activity

The total gamma activity from radioisotopes of fission products and induced activities shall not exceed 2.5 $\mu\text{Ci/gU}$. The gamma activity from individual radionuclides shall not exceed the following:

<u>Radionuclide</u>	<u>Maximum Gamma Activity $\mu\text{Ci/gU}$</u>
Cerium	1.0
Ruthenium	1.0
Cesium	0.2
Zirconium-Niobium	0.2
Any other individual radionuclide	0.1

3.1.2 Alpha Activity

The total alpha activity from neptunium and plutonium shall not exceed 0.1 $\mu\text{Ci/gU}$.

3.2 Uranium Metal

- 1) The total gamma activity from radioisotopes of fission products and induced activities shall not exceed 0.5 $\mu\text{Ci/gU}$. The gamma activity from individual radionuclides shall not exceed the following:

<u>Radionuclide</u>	<u>Maximum Gamma Activity $\mu\text{Ci/gU}$</u>
Cerium	0.2
Ruthenium	0.2
Cesium	0.05
Zirconium-Niobium	0.05
Any other individual radionuclide	0.05

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UNH Specifications for HM Solution to Y-12
(use only those within box)

The specification goals used by the SRP for the low enriched material are generally those given in Notice ORUF6CONV-71-1 dated 3/22/71 (Table 16). The exception is total alpha d/m/g U which was changed from 15,000 to 1,500 to 3,000 d/m/g U by DOE-OR (see page 12 for more details on this change). Except for the alpha specification, the analyses performed by the SRP are used only as SRP operating guides and not to make go/no-go shipping decisions.

d. Process Control and Potential Improvements

The HM process has been run at SRP since 1959. Many improvements have been made in both the process and the equipment since that time. The process is under rigid control with samples being taken of the product, all waste streams, and intermediate steps on a regular basis. Any significant improvement in product purity would have to be achieved through addition of a costly purification cycle. The Task Force judged this action would not be justified by any significant benefit.

2. Hanford Recycle Operations

a. Plant Description

In 1952, the U-Plant at Hanford began operating to recover uranium from the waste tanks for recycle, which was performed through 1958. In the Hanford REDOX and PUREX plants, processing technology permitted recovery of a uranium product (UNH). Recycle uranium as UNH from these two reprocessing facilities was sent to the uranium oxide plant (UO₃ Plant), which was part of the U-Plant complex, for conversion to UO₃ powder destined for offsite shipment.

Uranium was recycled from the REDOX plant from 1951 until 1967 when REDOX was shut down. PUREX recovered uranium from 1956 to 1972 when it was placed in standby. The plant resumed operation in November 1983, and UO₃ product shipments to the FMPC resumed in the spring of

Table 16

Item	Uranium Tetra- fluoride (UF ₄)	Numerical Value for Each Chemical Form*		
		Oxides** (Either UO ₂ , UO ₃ or U ₃ O ₈)	Uranyl Nitrate Crystals** (UO ₂ (NO ₃) ₂ · 6H ₂ O)	Uranyl Nitrate Solution (UO ₂ (NO ₃) ₂ + H ₂ O)
Maximum total beta activity from fission products as percent of the beta activity of aged natural uranium:				
a. provided that no less than 75% of the total fission product beta activity is due to ruthenium isotopes	100	100	200	200
b. provided that less than 75% of the total fission product beta activity is due to ruthenium isotopes	100	100	100	100
Maximum alpha activity from all transuranic elements in disintegrations per minute per gram of total uranium	15,000	15,000	15,000	1,500

*Numerical value given only where applicable

**Uranium tetra fluoride, uranium oxides and uranyl nitrate crystals shall be free-flowing powders

***See section 2, Packaging, of this notice

Depleted UNH Solution Specifications

1984. All recycle uranium from Hanford was sent to the FMPC as UO_3 product, except for the period 1959 to 1962 when UO_3 was shipped directly from Hanford to the Paducah Feed Plant. It should be noted that unirradiated uranium scrap from the N-reactor fuel manufacturing process is also returned to the FMPC for recovery. Radioactive contaminants in this material are unchanged from the as-received levels in the billets delivered from RMI.

There have been nine different production reactors which have operated at the Hanford site; uranium has been recovered and recycled from fuel that was irradiated in each of these reactors. Currently, the Hanford N Reactor is the only operational reactor at the site, and is the source of irradiated fuel for the PUREX and UO_3 facilities.

The block diagram (Figure 6) of the Hanford PUREX/ UO_3 facilities shows the recycle uranium flow and sample points in simplified form. Uranium product from the PUREX Plant is sampled in Tank K6 (uranium product sample tank) and in the UNH product storage tanks at PUREX prior to shipping to the UO_3 Plant via tank truck. Uranium oxide product is again sampled before shipment to NLO.

b. Contamination Control/Radiation Protection Practices

The contamination control and personnel protection practices at the Hanford UO_3 Plant are based on protection from potential hazards associated with uranium handling rather than from the radioactive contaminants in the uranium which are considered low enough to pose no unique safety hazards. The uranium recycle product is sampled twice before leaving the PUREX facility and must be within specification before it is sent to the UO_3 Plant. The controls are based on contaminant levels no greater than the maximum permitted in the specifications.

Personnel dosimetry and examination (whole body counting and urinalysis) have shown no detectable plutonium contamination in UO_3 Plant workers.

HANFORD RECYCLE URANIUM

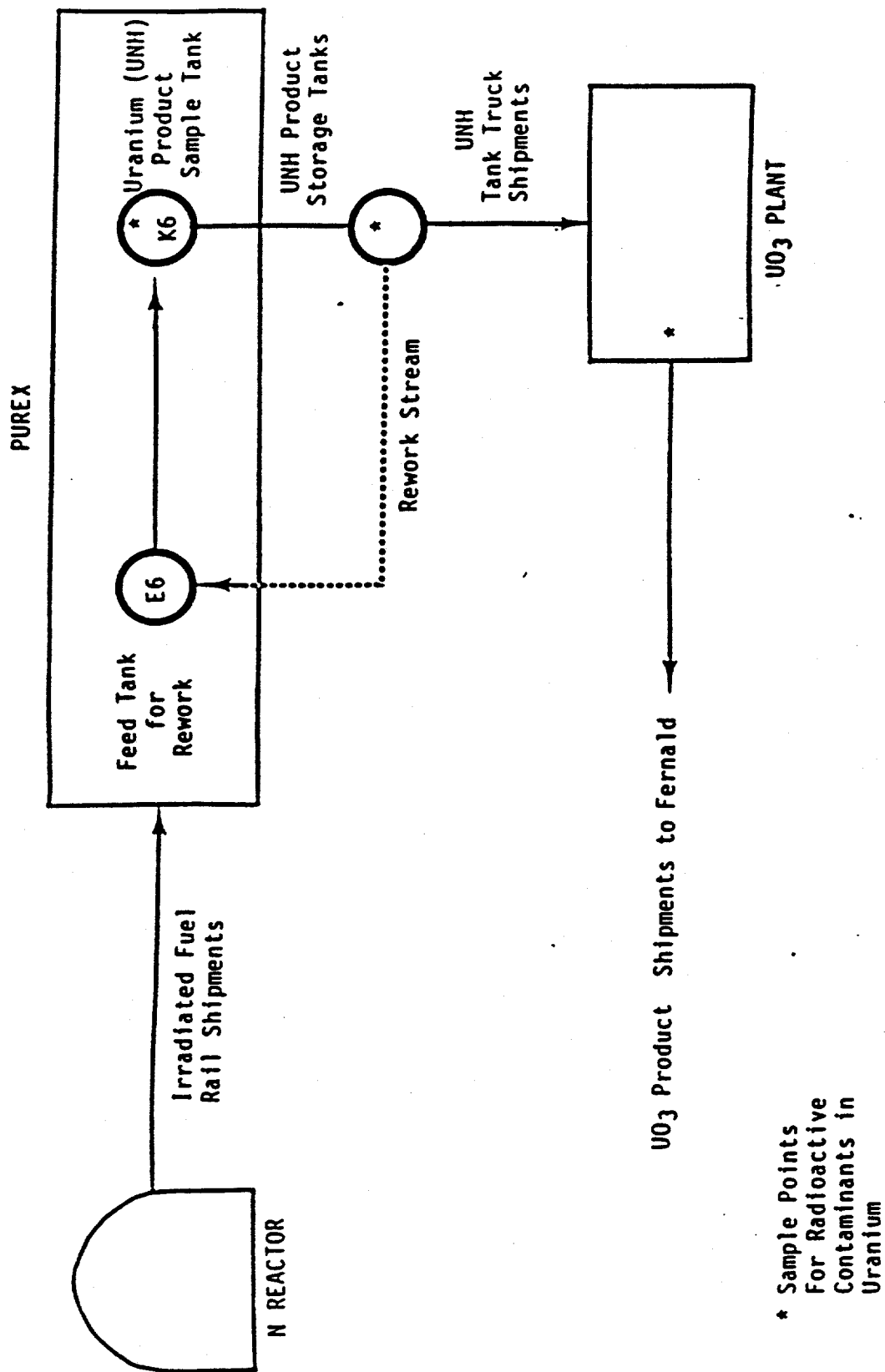


Figure 6
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c. Standards/Specifications

The specifications for Hanford recycle uranium are established for UNH product from the PUREX Plant (Table 17) and for UO_3 product from the UO_3 Plant (Table 18). These two sets of specifications are either identical for radioactive contaminants, or the specification required of PUREX UNH is lower (ZrNb-95, Ru-103 and RuRh-106) than the allowable contaminant limit in the UO_3 Product. Prior to the first offsite shipment of UO_3 product, a specification for allowable plutonium contamination was established at less than ten ppb per uranium. The specification remained in effect for all offsite shipments of Hanford recycle uranium, to the present. The current limit for plutonium contamination is a maximum of ten parts plutonium per billion parts uranium which is identical to the historical limit which apparently has existed since the origination of the uranium recycle effort at Hanford.

Uranium product is sampled for compliance with the specifications at three separate points:

1. Before PUREX UNH product is transferred to storage,
2. Before stored UNH product is transferred to the UO_3 Plant, and
3. Before UO_3 product is shipped to the FMPC.

If a batch of UO_3 product is outside of any of the nonradioactive specifications it is not shipped to the FMPC unless the prior written approval of the FMPC is given. UNH product from Purex that exceeds the specifications for any radioactive contaminants is not shipped to the UO_3 Plant.

1.0 INTRODUCTION

This document summarizes the criteria for PUREX Plant uranyl nitrate hexahydrate (UNH) and plutonium oxide (PuO_2) products. These criteria are currently contained in references 1 & 3-7 and are consolidated here for ready access and use.

2.0 PUREX UNH PRODUCT QUALITY CRITERIA

The criteria for PUREX Plant UNH product are included as Section 1.2-A of the reference 1, "Uranium Oxide Plant Specifications". Bases for these specifications are provided in reference 2. PUREX UNH not meeting the specifications must be approved in writing by Z-U-T-S Plants Process Control. Feed not meeting the uranium-235 requirement must be approved by Criticality Engineering and Analysis (CE&A).

Variable	Criteria
1. Uranium	3.9-4.4 lbs/gal
2. Nitric Acid	< 0.10 lb/gal
3. Actinides	< 1.0 wt% in uranium
a. Uranium-235	< 10 ppbp uranium
b. Plutonium	< 750 ppmp uranium
c. Thorium	
4. Fission Products	< 10 μ Ci/lb uranium
a. ^{95}Zr Nb	< 20 μ Ci/lb uranium
b. ^{103}Ru and $^{106}RuRh$	< 2 μ Ci/lb uranium
c. All others excluding ^{99}Tc	< 0.5 μ Ci/lb uranium average for each 480 tons
5. Metal Impurities	< 40 ppmp uranium
a. Iron	< 12 ppmp uranium
b. Nickel	< 16 ppmp uranium
c. Chromium	< 20 ppmp uranium
d. Sodium	
6. Organic	No visible organic phase

3.0 PUREX PuO_2 PRODUCT QUALITY CRITERIA

The criteria for PUREX PuO_2 are contained in references 3-7 which are reproduced in part in the following sections. Plutonium oxide with impurity levels exceeding the criteria may be accepted by the customer on an individual basis. Negotiations with the customer should be conducted before material with slightly high impurity levels is reworked.

The proposed specifications have been approved by the Nuclear Materials Processing and Scrap Management Task Force and will be issued as final and dated upon approval by the DOE area offices, Savannah River, Los Alamos, and DOE-ALO.

Variable		Limit	Violation Requirement	Difficulty	Safety	Product	Efficiency
C. <u>T-Hopper Containers, Loadout</u>							
1.	Powder Weight	< 6 tons			1		
D. <u>Product, UO₃ Powder Specifications</u>							
Analysis for the following quantities are performed prior to offsite shipment. Product not meeting the following specifications must be reported to the customer.							
1.	UO ₃ Assay	> 97%	HCR			1	
2.	Sulfur	250-400 ppmp uranium*	HCR			1	
3.	Actinides		HCR				
a)	Uranium-235	< 1.0 wt% in uranium				1	
b)	Plutonium	< 10 ppbp uranium					
c)	Thorium	< 750 ppmp uranium*					
4.	Fission Products		HCR				
a)	⁹⁵ ZrNb	< 15 µCi/lb uranium* < 10 µCi/lb uranium**				1	
b)	¹⁰³ Ru and ¹⁰⁶ RuRh	< 50 µCi/lb uranium* < 25 µCi/lb uranium**					
c)	All others excluding ⁹⁹ Tc	< 2 µCi/lb uranium* < 0.5 µCi/lb uranium**					
5.	Metal Impurities*	Rare Limits	HCR				
a)	Iron	40				1	
b)	Nickel	12					
c)	Chromium	16					
d)	Sodium	20					

* Values are based on a lot composite sample of 10 containers.

** Values apply to a 10-lot average.

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Task Force Observation

While the Hanford UO₃ product specifications are used as a working document by both the FMPC and Hanford, no written agreement between the two contractors exists to formalize the specifications.

The plutonium levels in UO₃ product from Hanford consistently have been below the 10 ppb level both historically and in present operations. Hanford analytical laboratory personnel indicate that the 10 ppb limit is approaching the detection limit for the analytical methods used to measure trace plutonium quantities. In the Hanford analytical laboratory 5 ppb is considered the practical detection limit, and a reported value of 1 ppb can be in error by 100 percent.

d. Process Control and Potential Improvements

The recycled uranium from Hanford is purified in the solvent extraction systems of the PUREX Plant and is recovered as a dilute UNH solution. In practice, the reported plutonium contaminant level in uranium product from PUREX since the plant restarted in 1983 has generally been reported lower than 5 ppb. In 1982, a brief preliminary review was made of methods that could be used to purify the PUREX uranium product further. This brief assessment showed that there might be ways of further decontaminating the uranium product; however, the techniques were unproven, complex, and would require long lead times for implementation.

3. Idaho Chemical Processing Plant (ICPP)

a. Plant Description

Since 1953, the ICPP has recovered uranium from spent (used) nuclear fuels, largely from government-owned reactors. The ICPP processes highly enriched (> 20% U-235) research, test reactor, and propulsion

reactors fuels. Presently, these fuels are aluminum, zirconium, or stainless steel clad and contain uranium alloys or uranium oxide mixed with other metals, metal oxides, or ceramic compounds. Special fuels or fuel materials occasionally are processed using customized processes and equipment in a hot cell facility.

The processing sequence begins when spent reactor fuels are dissolved in acid using one of several headend operations. This results in a solution containing uranyl nitrate hexahydrate (UNH). The UNH solution is then contacted with an organic solvent, tributyl phosphate (TBP). The uranium is extracted by the solvent while leaving most of the radioactive fission products and transuranics in the aqueous solution. Uranium is stripped from the solvent by water and extracted two more times with another organic solvent, methyl isobutyl ketone (hexone), for further purification.

The final product stream is UNH solution, practically free of fission products and other impurities. The uranyl nitrate solution is evaporated and denitrated to uranium trioxide (UO_3). The UO_3 product is mixed for homogeneity, sampled, and packaged. The packaged product is then shipped to other DOE facilities, such as the Y-12 Plant, for recycling in DOE programs.

b. Standards/Specifications

Purity specifications for UO_3 product produced by the ICPP is as follows:

<u>Element</u>	<u>ICPP Specifications (maximum ppm, normalized as U₃O₈)</u>
Aluminum	6000
Calcium	200
Chromium	200
Copper	200
Iron	1000
Magnesium	200
Sodium	200
Phosphorous	300
Silicon	250

Transuranic alpha activity <5000 dpm/g U

c. Potential Improvements

Significant quantities of the ICPP UO₃ product have been returned to the Y-12 Plant and the Portsmouth Oxide Conversion Facility for recycling in DOE programs. The Y-12 staff reported that early ICPP product averaged between 22 and 61 percent of Y-12's informal plutonium specification. It was also reported that early ICPP product was consistently greater than the specification on beta activity, averaging 472-510 percent of specification. Recent results (1977 to present) indicate the alpha ratio is about 31 percent of the Y-12 specification while the beta ratio is 68 percent of the specification. Since recent receipts from ICPP show an improvement in ICPP's ability to control product near both current ICPP production and Y-12 informal receipt specifications, the Task Force did not pursue additional process improvements with the ICPP.

4. The Paducah Feed Plant

When it was operational, the Paducah Feed Plant (shutdown in 1977) received uranium as UO₃ primarily from Hanford and converted this feed

material to uranium hexafluoride (UF_6) for eventual feed to the gaseous diffusion cascades. This was accomplished in three principal steps: reduction, hydrofluorination, and fluorination.

The reduction step was accomplished in the green salt (UF_4) plant using dissociated ammonia in two-stage fluidized bed reactors. In the second step, the UO_2 was hydrofluorinated to UF_4 by the addition of anhydrous hydrofluoric (HF) acid in three horizontal, screw type reactors arranged in series. The final step, the fluorination of UF_4 to UF_6 , was carried out in the UF_6 plant using elemental fluorine in tower reactors. Plant capacity was about 55-60 tons U per day with highly reactive UO_3 as feedstock.

It has since been concluded that the residual ash from the "bottoms" of the fluorination tower reactors tended to concentrate transuranic elements as a result of the "burning" process. The 22.5 metric tons uranium shipped to the FMPC in 1980 was this residual tower ash.

Task Force Observation

The Task Force was informed that the Paducah Plant still has an inventory of about 0.8 MTU as UO_3 and 1.2 MTU as miscellaneous solids in various containers that was recycled from the Hanford and/or SRP reactors. In addition, Paducah maintains an inventory of approximately 1.3 MTU contained in 117 ash receivers that remain from the cleanout campaign of the Paducah Feed Plant. It is expected that this 1.3 MTU would contain about the same level of transuranic contamination as that received by the FMPC in 1980 since this material represents the left-over heels. The Task Force offered a recommendation that a plan be developed for the safe disposition of this material.

5. The Portsmouth Oxide Conversion Facility

The Portsmouth Oxide Conversion Facility was used to convert high assay uranium compounds received from the ICPP (in the typical form of UO_3 or

U₃O₈) directly to UF₆. The facility used a pneumatic handling system, fluorine-oxide flame tower, and UF₆ transfer/trapping equipment, all of which were geometrically safe for processing highly enriched uranium. Although major components were contained within enclosures to prevent accidental contamination exposure to operating personnel and loss of material, the oxide conversion facility was not able to maintain adequate containment of the radioactive materials during operating periods. As such, the decision was made in the 1977 time period to shutdown the facility pending modifications to provide adequate containment measures. These modifications were never funded, and the facility has not been operated since.

Task Force Observation

An accurate determination of the quantity of recycle materials stored at Portsmouth was not made. However, the Task Force was told that several thousand 5-inch diameter cans possibly containing recycled UNH crystals exists at Portsmouth. The degree of contamination is not known. The Task Force was advised that Portsmouth has been sending non-UF₆ materials to either Y-12 (high assay) or the FMPC (low assay) for use as appropriate.

6. The DOE Gaseous Diffusion Plants

As part of its charter to estimate the amount of material possibly contaminated with transuranic and fission product elements in other areas, the Task Force learned that the GDPs are maintaining the following estimated inventories of UF₆ produced from reactor returns which contain transuranic and fission product elements:

Paducah	- 336 MTU
Oak Ridge	- 271 MTU
Portsmouth	- 6 MTU (some of this material is enriched to 86 percent U-235)

The Task Force recognized the need to address the cascade feeding of UF₆ produced from reactor returns. While the vast majority of the UF₆ that has been fed to the GDPs was produced from virgin uranium, some UF₆ recycle material obtained from reprocessed reactor fuel has been introduced to all three GDPs. The processing of uranium from spent reactor material has been intermittent and three distinct campaigns were noted: 1952-1964; 1969-1974; and 1976-1977. As indicated earlier in this report, virtually all of this recycle material came from the Savannah River and Hanford production reactors while very little was obtained from commercial power reactors.

Although reprocessing of spent reactor fuel was commercially banned in the United States during the Carter Administration, reprocessing continued in foreign countries. Today, DOE is contractually obligated to accept UF₆ produced from recycle material as feed to the enrichment complex if the material was originally enriched by DOE and if the UF₆ meets current feed specifications.

Over the years, recycle UF₆ has been fed to the GDPs. Because of this, several situations have been encountered as a result of the presence of trace concentrations of transuranics (primarily plutonium and neptunium) and fission products (primarily technetium). These contaminants gradually accumulated in the cascade, in waste streams, and in other process streams. At Portsmouth, for example, the accumulations of technetium in the upper sections of the cascade were such that special protective measures for maintenance workers were required to maintain exposures within safe limits.

The concerns posed by the feeding of recycle UF₆ as opposed to virgin UF₆ fall into three categories:

1. Transuranic elements
2. Fission products
3. Minor uranium isotopes

Each of these categories will be briefly discussed below. It should be recognized that the transuranic and fission product contaminants exhibit a similar behavior in that when introduced into the GDP, the contaminants are retained on process component surfaces. As their buildup continues, a point can be reached such that radiation activity levels can mandate special precautions to assure safe exposure levels to personnel.

The transuranic elements of interest (Pu-239 and Np-237) have been determined to deposit on plant equipment surfaces (Np-237) or be retained in the cylinders actually containing the recycle UF₆ (Pu-239). Data on transuranic contamination of enrichment plant equipment surfaces indicate that most of the contamination is from Np-237. As the levels of Np-237 increase, a point can eventually be reached where equipment handling could become a problem. The recently completed cascade improvement program effectively cleaned out the cascades. Relative to Pu-239, previous experience has shown that plutonium concentrates primarily in the UF₆ cylinder heel. One could therefore conclude that plutonium input to the enrichment plant will not reach a level requiring extensive personnel protection measures. However, one would need to address the disposal of residues from cylinder cleaning operations.

Fission products of primary interest are Tc-99, Ru-103, Ru-106, Sb-125, and Zr-95. Past experience with Tc-99 shows that technetium fluorides entering the cascade will in time be transported throughout the cascade and be partially removed with product and through purge cascade operations. Technetium must be controlled to avoid potential personnel internal exposure problems through inhalation. The remaining fission products of interest have been determined to accumulate in relatively small areas of the enrichment plant around the feed point. After large quantities of recycle UF₆ have been fed, significant radiation fields can develop thus presenting personnel hazards.

Minor uranium isotopes refer to U-232, U-233, U-236 and U-237. U-232 presents health hazards associated with handling uranium materials and maintaining plant equipment. U-233 poses a potential increase in alpha

radioactivity of uranium deposits in plant equipment. U-236 is a parasitic neutron absorber in nuclear reactors; as a result, it is necessary to increase the feed and separative work requirements to compensate for this parasitic effect in nuclear power reactors. U-237 decays rapidly to Np-237, thus the same effects from Np-237 as discussed previously apply.

Over the past few years, it has been DOE's policy to slowly batch feed recycle UF₆ into the Oak Ridge GDP. However, with the decision to place the Oak Ridge GDP in standby, DOE will be required to reexamine the strategy on where and when the recycle UF₆ will be disposed.

Over this time period, the GDP contractors have studied potential technology for removing transuranic and fission product contaminants from recycle UF₆. The basic premise is that feedstream trapping should be employed to remove these impurities before they enter the cascade. The review of existing technology shows that if one or, at most, two contaminants are present in the feedstream, trapping technology is adequate if safety limits are not made more restrictive. However, it is not possible at this time to specify a single acceptable trapping flow sheet design with reasonable confidence to handle all potential incoming contaminants of concern. Adverse chemical reactions between various feed contaminants and specific trapping agents make concrete recommendations on flow sheet design premature. More research, development and demonstration is necessary to address (1) trapping agent preparation, (2) in plant testing of contaminant trapping, (3) determining material balances for contaminants, (4) reduction of Ru-106 to assure safe operating levels (proven trapping technology for low concentrations of Ru-106 is not available), (5) consulting activities such as trap startup, operation, and disposal and cascade monitoring.

It is likely that a combination of revised UF₆ feed specifications (these specifications, currently under consideration by DOE, would significantly reduce the permissible level of fission products and transuranics in UF₆) and the implementation of chemical trapping will be necessary to permit the future introduction of recycle UF₆ to the GDPs.

Task Force Observation

In recent years, the GDP staffs have studied the problems associated with feeding recycle material to the GDPs. Currently, technology is available for removing certain transuranic and fission product contaminants from incoming feed. The most promising technology involves the feedstream trapping of the contaminants before they enter the cascades. Much work has been undertaken in this program. However, further efforts are needed to address a number of technical uncertainties.

III. SUMMARY AND CONCLUSIONS

Based on an analysis of the findings and observations of this report, the Task Force developed the following remarks.

Generic (Applies to All Sites)

- o A formal, technically sound, understood and accepted specification for maximum transuranic and fission product contaminants in uranium recycle material has probably never existed either within or between sites. Although most sites had their own "working" specification, there simply was no understanding and agreement on specifications for recycle material shipped to or from the DOE sites studied by the Task Force that had been agreed to, signed, and used for decision-making.
- o The Task Force found no reasons to propose a change or modification to the basic process flowsheets currently used at each of the plants reviewed. However, there does appear to be some instances where management attention is needed to improve environment, safety and health activities along with a stronger emphasis on "as low as reasonably achievable" (ALARA) concepts.

FMPC

- o The FMPC has not been required by DOE to maintain accountability records of transuranic and fission product elements in the quantities generally received by the FMPC. As such, the Task Force could not determine, with confidence, the quantity of contaminants that may have been received and processed at the FMPC. Only best estimates were available for the review.
- o Of all the plutonium estimated to have been received by the FMPC over the past 24 years (since plant startup), 50 percent of the plutonium was thought to have been contained in one shipment of Paducah Feed Plant ash in 1980. About 32 percent is believed to have been received from the Hanford site. The balance of the plutonium came from NFS-West Valley, the SRP, and other miscellaneous sources.

- o In June 1980, 22.5 metric tons of uranium ash from the Paducah Feed Plant was shipped to the FMPC for processing. This material was shipped to FMPC with DOE approval; however, it could not be established by the Task Force that DOE was formally aware of the plutonium levels in the ash. This recycle material, based on limited, non-homogeneous sampling, is estimated to have had plutonium levels ranging from 67 ppb to 7,757 ppb. In 1982, some of this material was processed and shipped to meet customer requirements. About 168 MTU of the remaining material (produced by diluting the original 22.5 MTU of Paducah Ash) currently remains at FMPC in the form of UO_3 with a maximum concentration of 43 ppb plutonium. Special precautions will need to be taken to process this material.
- o Overall, based on information made available, the Task Force found no evidence that DOE environmental, safety, and health guides had been exceeded nor was there any evidence that the health and safety of FMPC workers or the general public had been compromised due to operations involving recycle material. In addition, there was no evidence that the environment surrounding the FMPC had been adversely impacted from recycle operations. Nevertheless, deficiencies in the management control systems were noted which indicate a need for more environment, safety and health attention to uranium recycle processing operations.
- o The Task Force concluded that insufficient effort and attention was given to worker safety and radiation exposure control. For example, during routine operations the decision on whether an ingestion of radioactive material has taken or could take place rests with the worker. It did not appear to the Task Force that workers have had enough training and/or knowledge to intelligently make such decisions.
- o Routine processing of recycle materials containing less than 10 ppb plutonium can be accomplished with existing administrative and radiation protection practices. This is true since uranium is the dominant radionuclide for health protection purposes at a plutonium concentration less than 10 ppb. Most of the radiation protection practices are those that should be

implemented to support uranium operations. The requirement should be to assure that uranium is always the controlling nuclide for any processing operation regardless of the contaminants that may be in the feed material.

- o Deficiencies in the FMPC Health Physics and Environmental Programs were noted by the Task Force. However, recent special reviews have pointed out these deficiencies, and as such, the Task Force chose not to repeat those same findings and recommendations previously observed.

RMI

- o Depleted and slightly enriched uranium ingots from FMPC are extruded into tubes at RMI and then shipped to Hanford or back to the FMPC for final work. No contaminants are added to or removed from the uranium being worked at RMI. RMI does, however, convert uranium turnings to an oxide form prior to being returned to FMPC.
- o RMI is not equipped to perform sampling analysis for transuranic or fission products. RMI analyses are generally accomplished by the FMPC. In addition, outside laboratories and expertise are available to RMI.
- o A recent risk and vulnerability study of RMI outlined several improvements that would benefit recycle material processing operations at RMI.

Y-12

- o Limited data exists at Y-12 on the transuranic and fission product content of recycle material receipts, processing streams, and product streams. As in the case of the FMPC, Y-12 has not been required to maintain accountability data on plutonium, other transuranic elements, or fission products.
- o An exposure assessment of recycle workers indicate these workers have an external exposure 1.2-1.6 times the exposure of Y-12 workers handling unirradiated (virgin) uranium materials. The internal dose to the recycle

workers were calculated by Y-12 to be 0.019 rem per year (committed dose to the bone) per employee. The Task Force agreed with Y-12 that these exposures do not represent a significant health or safety risk to the recycle workers. These calculations were based on a review of employee exposure records.

- o Based on limited sampling since 1977, the Y-12 staff has noted a buildup of fission products in both the liquid and solid waste streams as a result of processing recycle material. It appears that this waste stream buildup has taken place as the fission product concentration in Y-12 product has decreased. The waste streams were previously routed to the S-3 Ponds; however, the S-3 Ponds have been closed, and a closure plan has been developed by the Y-12 Plant in conjunction with local, state and federal agencies.

Paducah Feed Plant

- o The Paducah Feed Plant (shutdown in 1977) maintains an inventory of about 0.8 MTU as UO_3 and 1.2 MTU as miscellaneous solids processed from recycle material. In addition, about 1.3 MTU as ash remains from the cleanout of the Paducah Feed Plant. Plans for disposition of this recycle material have not been formulated. Such a plan is a recommendation of the Task Force.

Portsmouth Oxide Conversion Facility

- o This facility (shutdown since about 1977) was used to convert high assay uranium recycle material to UF_6 for feed into the gaseous diffusion plant. An estimated several thousand 5-inch diameter cans possibly containing some level of transuranic and fission product contaminants are in storage at the site. Non- UF_6 material is being shipped to Y-12 (high assay) or the FMPC (low assay) on an as-needed basis.

Gaseous Diffusion Plants (GDP)

- o It is estimated that the three GDPs currently have about 613 MTU as UF_6 that was commercially processed from recycle material. The assay of this material generally is about 0.8 percent U-235, but some material goes as high as 86 percent U-235. In recent years, the problems associated with feeding uranium recycle material to the GDPs have been studied. The technology for removing transuranics (primarily Np-237) and fission products (primarily Tc-99) from incoming feed is judged to be available; however, this technology has not been demonstrated on a production level. Action on this technology is currently receiving low priority attention by DOE due to economic considerations.

IV. RECOMMENDATIONS

Recommendations

The Uranium Recycle Task Force offers the following recommendations for DOE's consideration:

Generic (Applies to All Sites)

Mutually agreeable and technically sound transuranic and fission product element specifications should be established between shipper and receiver for all recycle material shipped to and from all DOE sites handling recycle material. The specifications should be agreed upon and signed by all involved contractors, and approved by the appropriate DOE field offices. These specifications should be provided to all affected plants.

To support the recycle material specifications noted above, a documented technical basis should be prepared for those radioactive contaminants (transuranics and fission products) covered by the specification. A justification for each limit should be given. To implement this recommendation, DOE established a multi-contractor Specifications Task Group to develop these specifications under the auspices of the Uranium Recycle Task Force.

FMPC

FMPC management should:

1. Continue to recognize the 10 ppb Pu (on a uranium basis) specification (upper limit) on recycle materials until the results of the Specifications Task Group noted above completes its work. The 10 ppb Pu guide is based on the rationale given on pages 16-19 of this report.

2. Regarding any remaining (especially the 168 MTU from the Paducah Ash) or future receipts of recycle material at FMPC, formalize the operating and environment, safety and health procedures; handling methods; and analyses to be utilized in future processing operations which will clearly demonstrate that no adverse environmental, safety or health effects will occur from recycle material processing. To the extent possible, engineering controls are preferred over administrative controls. The processing of recycle material in concentrations greater than 10 ppb plutonium will require special DOE-ORO approval if it is necessary to process recycle material prior to completion of the Specification Task Group work.
3. Comprehensively review the analytical and radiochemical procedures and control programs for determining transuranic and fission product elements in the incoming, in-process, and product streams as well as in environmental samples. Procedures and equipment should be upgraded to state-of-the-art technology and be capable of identifying individual radionuclides, e.g., Pu-238 and Pu-239 to the extent possible.
4. Obtain expert advice and guidance in establishing a bioassay program including in-vivo (whole body) counting for FMPC workers. To the extent feasible, the program should include the measurement of transuranic and fission product burdens. Until such time as an onsite capability can be established, FMPC should obtain bioassay data (if this has not already been accomplished) on those workers involved in recycle material operations. In addition, those recycle workers with the highest potential of lung internal deposits of plutonium should be sent to facilities with in vivo counters for measurement.
5. Review the FMPC training program for operators, supervisors and maintenance personnel to assure compliance with DOE Order 5480.1A, Chapter V, paragraph 8. In addition, FMPC should review operating and environment, safety and health (ES&H) procedures to assure these procedures are current, readily available to and used by personnel, periodically reviewed/updated as necessary, and have been properly reviewed by the ES&H staff.

RMI

RMI management should:

1. Recognize and support the FMPC 10 ppb Pu (on a uranium basis) specification on DOE recycle materials until the results of the Specifications Task Group noted above completes its work. The 10 ppb Pu guide is based on the rationale presented on pages 16-19 of this report.
2. Implement a reasonable program of sampling and analyses on incoming, in-process and outgoing batches of recycle material. RMI efforts should include process residues and waste streams, as appropriate.
3. Include all uranium handling workers in the RMI bioassay program if this has not already been accomplished. To the extent feasible, the bioassay program should include transuranic and fission product measurements.

Y-12

Y-12 management should:

Review radiochemical procedures and control programs associated with the analysis of uranium recycle processing operations.

Paducah Feed Plant

Paducah management, in conjunction with DOE, should:

Conduct an exposure assessment (to transuranics and fission products) for those workers involved in the processing of recycle materials at the Paducah Feed Plant.

Portsmouth Oxide Conversion Facility

Portsmouth management, in conjunction with DOE, should:

Conduct an exposure assessment (to transuranics and fission products) for those oxide conversion facility workers involved in the processing of recycled materials.

DOE Gaseous Diffusion Plants (Oak Ridge, Paducah and Portsmouth)

GDP contractor management should:

Provide DOE with a report on the options available and recommendations for a safe, technically sound, and feasible method for disposition of current inventories and future GDP receipts of uranium recycle material (UF₆ and non-UF₆). The report should include, as a minimum, (a) an assessment of the problems presented by recycle materials on the GDP complex, (b) available disposition options, and (c) a recommended course of action to DOE. DOE will be responsible for a final decision in this matter.